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No. 95-728

Supreme Court, U.S.
FILED

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IN THE
Supreme Court of the United States

OCTOBER TERM, 1995

WARNER-JENKINSON COMPANY, INC.,
Petitioner,

v.

HILTON DAVIS CHEMICAL CO.,
Respondent.

On Writ of Certiorari to the
United States Court of Appeals
for the Federal Circuit

JOINT APPENDIX

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**U.S. DISTRICT COURT
FOR THE SOUTHERN DISTRICT OF OHIO
(CINCINNATI)**

Civil Docket for Case #: 91-CV-218

HILTON DAVIS CHEM

v.

WARNER-JENKINSON

DOCKET ENTRIES

Date	No.	PROCEEDINGS
4/1/91	1	Complaint, with Patent attached
4/23/91	2	Answer

4/1/92	21	Warner-Jenkinson's Motion for Summary Judgment of Non-Infringement

5/11/92	50	First day of trial

5/19/92	59	Fifth day of trial; Warner-Jenkinson's Motion for Summary Judgment of Non-Infringement Denied

6/16/92	87	Special Verdict

6/17/92	89	Motion for Permanent Injunction
6/22/92	90	Judgment

Date	No.	PROCEEDINGS
6/22/92	152	Corrected Judgment

7/1/92	92	Warner-Jenkinson's Motion for Judgment as a Matter of Law

10/20/92	150	Order for Permanent Injunction
10/20/92	151	Order Denying Warner-Jenkinson's Motion for Judgment as a Matter of Law

11/16/92	154	Notice of Appeal
11/17/92	155	Amended Notice of Appeal

**U.S. COURT OF APPEALS
FOR THE FEDERAL CIRCUIT**

Federal Circuit Docket for Case #: 93-1088

HILTON DAVIS CHEMICAL Co.,
Plaintiff-Appellee

v.

WARNER-JENKINSON COMPANY, INC.,
Defendant-Appellant

DOCKET ENTRIES

Date	No.	PROCEEDINGS

7/9/93	10	Oral Argument Before the Panel

12/3/93	15	Order that Appeal Shall be Decided <i>in banc</i> and Setting Forth Issues to Address

3/9/94		Oral Argument <i>in banc</i>

8/9/95	48	Per Curiam <i>in banc</i> Opinion Affirming as to Issue of Infringement With Separate Concurring and Dissenting Opinions; Panel Opinion Affirming as to Validity

UNITED STATES COURT OF APPEALS
FOR THE FEDERAL CIRCUIT

93-1088

HILTON DAVIS CHEMICAL Co.,
Plaintiff-Appellee,

v.

WARNER-JENKINSON COMPANY, INC.,
Defendant-Appellant,

ORDER

The appeal, having been heard by a panel of the court and, thereafter, a majority of circuit judges in regular active service having acted sua sponte in accordance with 28 U.S.C. § 46(c) and Fed. R. App. P. 35(a),

IT IS ORDERED that the appeal shall be decided in banc.

IT IS FURTHER ORDERED that the parties shall file supplemental briefs on the following schedule:

The brief for appellant on rehearing in banc shall be filed on or before January 7, 1994. The brief for appellee on rehearing in banc shall be filed on or before February 4, 1994.

The following questions only shall be addressed in the briefs:

(1) Does a finding of patent infringement under the doctrine of equivalents require anything in addition to proof of the facts that there are the same or substantially the same (a) function, (b) way, and (c) result, the so-called triple identity test of *Graver Tank v. Linde Air*

Products Co., 339 U.S. 605, 85 USPQ 328 (1950), and cases relied on therein? If yes, what?

(2) Is application of the doctrine of equivalents by the trial court to find infringement of the patentee's right to exclude, when there is no literal infringement of a claim, discretionary in accordance with the circumstances of the case?

(3) Is the issue of infringement under the doctrine of equivalents an equitable remedy to be decided by the court, or is it, like literal infringement, an issue of fact to be submitted to the jury in a jury case?

Amicus curiae briefs on rehearing in banc may be filed in accordance with Rule 29.

Oral argument will be scheduled after the briefs have been filed.

FOR THE COURT

/s/ Francis X. Gindhart
FRANCIS X. GINDHART
Clerk

12/3/93

cc: J. Robert Chambers, Esq.
David E. Schmit, Esq.

[Filed Dec. 3, 1993]

[Filed Apr. 1, 1991]

UNITED STATES DISTRICT COURT
SOUTHERN DISTRICT OF OHIO
WESTERN DIVISION

Civil Action No. _____

HILTON DAVIS CHEMICAL Co.,
vs. *Plaintiff,*

WARNER-JENKINSON,
_____ *Defendant.*

COMPLAINT FOR PATENT INFRINGEMENT
AND JURY DEMAND

Plaintiff, complaining of defendant, alleges as follows
through its undersigned attorneys:

FIRST CAUSE OF ACTION

1. This cause of action is for infringement of a United States patent and arises under the patent laws of the United States, Title 35 of the United States Code. The Court has jurisdiction of this cause of action under said Title 35 and under 28 U.S.C. § 1338. Venue is proper in this district pursuant to 28 U.S.C. §§ 1391(c) and 1400(b).

2. Plaintiff is a corporation duly organized and existing under the laws of the State of Delaware and having its principal place of business at 2235 Langdon Farm Road, Cincinnati, Ohio 45237.

3. Upon information and belief, Defendant has its place of business at 2526 Baldwin Street, St. Louis, Missouri 63106.

4. On December 24, 1985, United States Patent No. 4,560,746 was duly and legally issued for the process of the Ultrafiltration Process for Purification of Dyes Useful in Food Stuffs. Said patent is owned by the Plaintiff.

A copy of United States Patent No. 4,560,746 is attached hereto as Exhibit A.

5. Defendant has willfully infringed United States Patent No. 4,560,746.

WHEREFORE, Plaintiff demands judgment as follows:

(a) that Defendant, its officers, agents, servants, employees and attorneys and all persons in active concert with them, or any of them, be preliminary and permanently enjoined from infringing United States Patent No. 4,560,746;

(b) that Plaintiff be awarded damages adequate to compensate for Defendant's infringement, said damages to be trebled because of the willful nature of Defendant's said acts; and

(c) that Plaintiff have such other and further relief as this Court may deem just and proper, together with reasonable attorneys' fees and the cost and disbursements of this action.

Plaintiff demands a trial by jury.

HILTON DAVIS CHEMICAL Co.

By: /s/ David E. Schmit
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United States Patent [19]

Rebhahn et al.

[11] Patent Number: 4,560,746

[45] Date of Patent: Dec. 24, 1985

[54] ULTRAFILTRATION PROCESS FOR PURIFICATION OF DYES USEFUL IN FOODSTUFFS

[75] Inventors: Robert W. J. Rebhahn, Berkley, Mass.; Wayne L. Cook, Cincinnati, Ohio

[73] Assignee: The Hilton-Davis Chemical Co., Cincinnati, Ohio

[21] Appl. No.: 677,118

[22] Filed: Nov. 30, 1984

Related U.S. Application Data

[63] Continuation-in-part of Ser. No. 481,038, Mar. 28, 1983, abandoned.

[51] Int. Cl.⁴C09B 29/16; C09B 67/54;
C09B 69/00[52] U.S. Cl.534/840; 534/554;
534/883; 534/884; 534/887[58] Field of Search260/208, 144 P; 534/887,
573, 840, 883, 884; 546/174

[56] References Cited

U.S. PATENT DOCUMENTS

3,249,444 5/1966 Bollenback et al.
 3,544,455 12/1970 Adams et al.
 4,165,288 8/1979 Teed et al.
 4,189,380 2/1980 Booth et al.260/144 X

FOREIGN PATENT DOCUMENTS

59782 9/1982 European Pat. Off.
 816730 10/1981 South Africa

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Colour Index, 3rd Edition, vol. 4, p. 4087, #15985, (1971).

Osmonics, Inc. Bulletin No. 109, Mar. 1978.

Spatz I, D. Dear., Reverse Osmosis/Ultrafiltration Application to Water Reuse and Material Reclamation, 5-1-75.
 Spatz II, D. Dean, Industrial Wastes, Jan.-Feb. 1974, pp. 20-24.

Primary Examiner—Floyd D. Higel

Attorney, Agent, or Firm—William G. Webb; B. Woodrow Wyatt; Paul E. Dupont

[57] ABSTRACT

The disodium salt of 1-[(6-methoxy-4-sulfo-3-methylphenyl)azo]-2-naphthol-6-sulfonic acid (FD and C Red 40), the disodium salt of 1-[(4-sulfophenyl)azo]-2-naphthol-6-sulfonic acid (FD and C Yellow 6), the trisodium salt of 1-[1-(4-sulfonaphthyl)azo]-2-naphthol-3,6-disulfonic acid (FD and C Red 2), the disodium salt of 2-[1-(4-sulfonaphthyl)azo]-1-naphthol-4-sulfonic acid (Carmoisine) and the sodium salt of 2-(2-quinolyl)-1,3-indanedione-sulfonic acid (D and C Yellow 10) are prepared and purified in high yield and in a high state of purity by subjecting their aqueous reaction mixtures to ultrafiltration through a membrane of such structure and under such conditions that the impurities are separated from the reaction mixtures, and the products are concentrated in high purity concentrates from which the products can be isolated directly by evaporation of the solvent.

17 Claims, No Drawings

ULTRAFILTRATION PROCESS FOR PURIFICATION OF DYES USEFUL IN FOODSTUFFS

RELATED APPLICATION

This is a continuation-in-part of our prior, copending application Ser. No. 481,038, filed Mar. 28, 1983, now abandoned.

BACKGROUND OF THE INVENTION

(a) Field of the Invention

This invention relates to the field of purification, by ultrafiltration techniques, of dyes useful in foodstuffs.

(b) Information Disclosure Statement

Bollenback et al. U.S. Pat. No. 3,249,444, patented May 3, 1966, describes an ultrafiltration process for increasing the tinctorial power of caramel color in which sugar, i.e. uncaramelized sugar, is separated from caramel color by ultrafiltration through a semi-permeable membrane which permits passage of small, uncolored molecules in solutions containing caramel color and rejects the passage of larger, polymeric caramel color molecules, thus enhancing the color of the concentrate. Preferred membranes for the process are made of vinyl plastics, and preferred pressures are in the range from 20 to 100 p.s.i.g.

Adams et al. U.S. Pat. No. 3,544,455, patented Dec. 1, 1970, discloses a process for the purification of itaconic acid by reverse osmosis through a semi-permeable membrane composed of cellulose acetate or polyamide in which itaconic acid and water are forced to the downstream side of the membrane, while inorganic salts, colored materials and organic materials remain on the upstream side. The process is carried out under a hydrostatic pressure of from

100 to 1,000 p.s.i.g. and at a pH in the range from 2 to 4.

Teed et al. U.S. Pat. No. 4,165,288, patented Aug. 21, 1979, discloses a process for the concentration and partial purification of textile vat dyes for recovery and re-use of the same by subjecting the dye solutions from dyeing operations to reverse osmosis through a semi-permeable membrane, impurities being collected in the permeate and the dye being concentrated in the concentrate. The process is carried out at hydrostatic pressures from 400 to 1,300 p.s.i.g. and at temperatures from 130° F. to 212° F. In order to prevent plugging of the membrane, a turbulent flow of liquid is needed.

EPO Application No. 59,782, published Sept. 15, 1982, discloses a process for concentration, to unspecified levels of purity, of certain anionic dyes, useful in the printing and dyeing of synthetic fibre materials, by passing solutions or suspensions of the dyes through a semi-permeable membrane with a pore diameter of 1-500 Angstroms.

South African Pat. No. 81/6,730, patented Sept. 6, 1982 discloses a process for the preparation of concentrated solutions of anionic dyes, of unspecified purity and useful in printing inks and dye baths, comprising passing a suspension or solution of the dye over a semi-permeable membrane containing ionic groups and having a pore diameter of 1-500 Angstroms.

Osmonics, Inc. Bulletin No. 109 describes the use of a variety of reverse osmosis/ultrafiltration membranes for a variety of purposes, including use of Osmonics, Inc. SEPA-50 membrane in textile dye removal. The membrane is said to give 40-70% sodium chloride re-

jection and a molecular weight cut-off of approximately 600 for organic materials.

Spatz, Reverse Osmosis/Ultrafiltration Application to Water Reuse and Material Reclamation, May 1, 1975, at page 8, discloses that reverse osmosis/ultrafiltration membranes can be used to remove organic dyes and that some organic dyes are poorly rejected by the membrane. That is, the dye would pass through the membrane.

Spatz, Industrial Wastes, January/February 1974, pages 20-24, discloses the use of reverse osmosis/ultrafiltration membrane methods for concentrating sucrose/dye solutions used in Maraschino cherry processing so that used dyeing solutions, rather than being discarded as in the past, can be concentrated down and reused.

Thus although the general concept of the use of reverse osmosis/ultrafiltration techniques to purify and concentrate a variety of materials is known, so far as is known, the application of this technology to dyes has been restricted to its use for merely concentrating dyes for reuse either in the textile industry, as in Teed or EPO Application No. 59,782, or in Maraschino cherry dyeing, as in Spatz (Industrial Wastes).

SUMMARY

In accordance with the present invention, certain dyes useful in foodstuffs are not merely concentrated, as provided by the prior art, but rather are prepared in molar yields which are unprecedented in the food dye stuff industry, and at purity levels which exceed the purity standards required by the U.S. Food and Drug Administration.

These unprecedented results are achieved by essentially incorporating the purification of the dyes as part of a continuous preparation/purification process, the purification being effected by subjecting an aqueous solution of the reaction mixture resulting from preparation of the dye to ultrafiltration under conditions such that the dye can be isolated by evaporation of its solution in molar yields of approximately 98% and in a state of purity of approximately 90%. In certain instances molar yields as low as around 75% are obtained, but even in such

cases, the state of purity of the dyes which can be achieved by the present process is around 90%. In practicing the invention, it is preferred to subject the reaction mixture, which results from the preparation of the dyes, directly to ultrafiltration without isolation of the product. Alternatively, however, the products can be isolated in crude form from the reaction mixtures, either by salting out or by spray drying, and the crude product then redissolved in water and the solution subjected to ultrafiltration.

Accordingly, the invention comprises a process for purification of a dye selected from the group consisting of the disodium salt of 1-[(6-methoxy-4-sulfo-3-methylphenyl)azo]-2-naphthol-6-sulfonic acid, the disodium salt of 1-[(4-sulfophenyl)azo]-2-naphthol-6-sulfonic acid, the trisodium salt of 1-[1-(4-sulfonaphthyl)azo]-2-naphthol-3,6-disulfonic acid, the disodium salt of 2-[1-(4-sulfonaphthyl)azo]-1-naphthol-4-sulfonic acid and the sodium salt of 2-(2-quinolyl)-1,3-indanedione-sulfonic acid as the products of their respective preparations via coupling of diazonium salts, in the case of the first four named dyes, and via sulfonation, in the case of the last named dye, where said dyes are present in the final reaction mixtures along with impurities, which process comprises subjecting an aqueous solution of the

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reaction mixture resulting from said coupling or said sulfonation to ultrafiltration through a membrane having a nominal pore diameter of from 5 to 15 Angstroms under a hydrostatic pressure of approximately 200 to 400 p.s.i.g. to thereby cause separation of the impurities into the permeate and concentration of the products in the concentrate.

DETAILED DESCRIPTION INCLUSIVE OF THE PREFERRED EMBODIMENTS

The dyestuffs, FD and C Red 40 and FD and C Yellow 6, chemically the disodium salt of 1-[(6-methoxy-4-sulfo-3-methylphenyl)azo]-2-naphthol-6-sulfonic acid and the di-

sodium salt of 1-[(4-sulfophenyl)azo]-2-naphthol-6-sulfonic acid, respectively, are approved by the U.S. Food and Drug Administration, and Amaranth and Carmoisine, chemically the trisodium salt of 1-[1-(4-sulfonaphthyl)azo]-2-naphthol-3,6-disulfonic acid and the disodium salt of 2-[1-(4-sulfonaphthyl)azo]-1-naphthol-4-sulfonic acid, respectively, are approved by the European Economic Community (E.E.C.), for use in foodstuffs, but as foodstuff additives, they must meet certain strict standards of purity. FD and C Red 40, FD and C Yellow 6, FD and C Red 2 and Carmoisine are each prepared in essentially "one pot" reactions by the diazotization of 5-methoxy-2-methylsulfanilic acid (FD and C Red 40), sulfanilic acid (FD and C Yellow 6), and sodium 4-amino-1-naphthalene sulfonate (FD and C Red 2 and Carmoisine), followed by coupling of the resulting respective diazonium salts with sodium 2-naphthol-6-sulfonate, for the preparation of FD and C Red 40 and FD and C Yellow 6, or with disodium 2-naphthol-3,6-disulfonate, for the preparation of FD and C Red 2, or with sodium 1-naphthol-4-sulfonate, for the preparation of Carmoisine. Moreover D and C Yellow 10, chemically the sodium salt of 2-(2-quinolyl)-1,3-indanedione sulfonic acid, is approved for use as a drug and cosmetic coloring agent. D and C Yellow 10 is also prepared in an essentially "one pot" procedure involving condensation of 2-quinaldine with phthalic anhydride followed by sulfonation of the resulting 2-(2-quinolyl)-1,3-indanedione.

Dyestuffs which are not intended for human consumption, for example those intended for use as textile dyes or printing inks, whose state of purity for such ultimate uses is not critical, can, of course, be isolated directly by evaporative concentration of the reaction mixtures in which they are produced followed by collection of the dye. Using such procedures, the final products are contaminated with major amounts of unpurities whose presence would not adversely affect the use of the dyes. However, in dyes used as food coloring agents, for ex-

ample, such product isolation procedures in which large amounts of impurities would be carried along with the product, would be completely unacceptable.

Therefore dyestuffs used as food coloring agents have conventionally been separated from impurities present in their reaction mixtures by crystallization. However, because the various dyes which are the subject of this invention are all moderately soluble in water, they have heretofore been purified of impurities present in reaction mixtures in which they are produced by the addition of large quantities of salt (sodium chloride) so as to "salt out" the product. However such salting out processes have several disadvantages. To begin with, the salt required is expensive, and furthermore the brine produced in the final filtrate presents a

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major disposal problem for industry. Moreover, because of the high solubility of the dyes, even in brine, a high percentage of the product (from around 12% to around 20%) is lost in the brine. Thus in a typical batch containing 2,500 pounds of FD and C Red 40 in a final reaction mixture, 13,200 pounds of salt would be required in order to recover about 2,200 pounds of product, the remaining 300 pounds being lost in the filtrate after collection of the solid product by filtration.

It will be seen then that, in view of the above circumstances, the cost of the salt, the added cost to industry of disposing of the brine and the cost of the lost product can, in toto, be very substantial, resulting in greatly increased costs of the products as sold. The novel method provided by the present process overcomes these disadvantages by avoiding the need for salt and by providing for recovery of up to 98% of the product actually produced in the reaction mixture. In addition, the method produces a product having a state of purity which exceeds the purity standards required by regulatory agencies,

such as the U.S. Food and Drug Administration, or by the European Economic Community.

In accordance with the present invention for the purification of FD and C Red 40, FD and C Yellow 6, FD and C Red 2 and Carmoisine, all prepared by coupling of an appropriate diazonium salt with a naphthol sulfonic acid derivative, therefore, these advantages are realized by incorporating the purification step as part of an essentially continuous preparation/purification procedure in which the reaction mixtures resulting from the coupling of the diazonium salts of 5-methoxy-2-methylsulfanilic acid (for FD and C Red 40), sulfanilic acid (for FD and C Yellow 6) or 4-amino-1-naphthalene sulfonic acid (for FD and C Red 2 and Carmoisine), in the form of the corresponding sodium salts in each case, with sodium 2-naphthol-6-sulfonate, disodium 2-naphthol-3,6-disulfonate or sodium 1-naphthol-4-sulfonate, as the case may be, are subjected directly to ultrafiltration through a membrane having a nominal pore diameter of such limiting size that the membrane will reject all molecules of a molecular size either the same as or greater than the products, FD and C Red 40, FD and C Yellow 6, FD and C Red 2 or Carmoisine, but which will allow passage of smaller molecules, including unreacted starting materials, i.e. 5-methoxy-2-methylsulfanilic acid (also known as cresidine sulfonic acid and hereinafter designated CSA), as used in the preparation of FD and C Red 40, sulfanilic acid (hereinafter designated SA), as used in the preparation of FD and C Yellow 6, 4-amino-1-naphthalene sulfonic acid, as used in the preparation of FD and C Red 2 and Carmoisine, sodium 2-naphthol-6-sulfonate (also known as Schaeffer's salt and hereinafter designated SS), disodium 2-naphthol-3,6-disulfonate and sodium 1-naphthol-4-sulfonate, as well as sodium chloride, which are the principal impurities to be found in the various aqueous product mixtures.

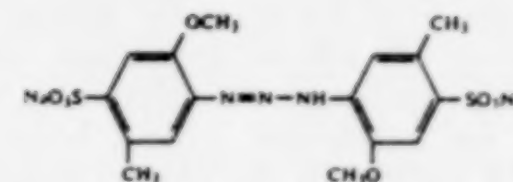
The reaction mixtures, however, may also contain impurities resulting from preparation of the 5-methoxy-2-

methylsulfanilic acid (used in the preparation of FD and C Red 40), sulfanilic acid (used in the preparation of FD and C Yellow 6) or 1-aminonaphthalene-4-sulfonic acid by sulfonation of the respective 5-methoxy-2-methylaniline, aniline or 1-aminonaphthalene. These latter impurities include higher sulfonates of 5-methoxy-3-methylaniline, aniline and 1-aminonaphthalene (hereinafter designated HS) and sodium sulfate).

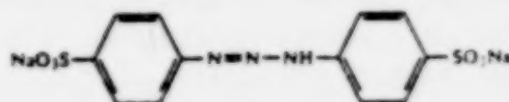
[5]

In the case of D and C Yellow 10, the principal impurities which may be found in the final product are the intermediate 2-(2-quinolyl)-1,3-indanedione, also known as Yellow 11, the disodium salt of 2-(2-quinolyl)-1,3-indanedione disulfonic acid, which results from disulfonation of Yellow 11, unreacted quinaldine, phthalic anhydride, an impurity of unknown structure designated chlorinated Yellow 11, which is produced during the high temperature, zinc chloride catalyzed condensation of 2-quinaldine and phthalic acid, and the chlorides and sulfates of sodium.

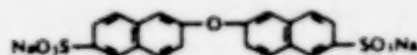
Moreover, the reaction mixtures may, in addition, contain in minor amount a variety of other impurities formed as undesired by-products, either in the preparation of FD and C Red 40 and FD and C Yellow 6, or in the preparation of starting materials used in their preparation. Thus the diazonium salt formed from 5-methoxy-3-methylsulfanilic acid can react with the amino nitrogen atom of undiazotized 5-methoxy-3-methylsulfanilic acid, in the preparation of FD and C Red 40, to form a triazene, which, in the form of the disodium salt, has the structure:



and is identified as the disodium salt of 4,4'-(diazoamino)-bis-(5-methoxy-2-methylbenzenesulfonic acid) (hereinafter designated DMMA); or the same type of triazene:



identified as the disodium salt of 4,4'-(diazoamino)-bis-(benzenesulfonic acid) (hereinafter designated DAAB) can be formed in the preparation of FD and C Yellow 6; or a dinaphthyl ether:



identified as the disodium salt of 6,6'-oxybis-(2-naphthalenesulfonic acid) (hereinafter designated DONS) can be produced as a by-product in the preparation of sodium 2-naphthol-6-sulfonate by sulfonation of 2-naphthol.

Thus it will be seen that the reaction mixtures resulting from the preparation of the various dyes purified in accordance with the invention can possibly contain in minor amounts, along with the desired products, a complex mixture of impurities. The economical separation of the wide variety of impurities from the products in the present process, in order to achieve the levels of purity required by FDA regulations, is thus a critical aspect of the preparation of these dyestuffs for commerce.

The presence of some of those impurities in the final products can, of course, be minimized by use of purified starting materials so that impurities from that source are

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not carried along in the synthetic process to the final product mixture. It will be appreciated from the foregoing that large molecular size impurities, which will be retained by the membranes, cannot be present in the solution to be purified by the present process at concen-

trations which would be unacceptable in the final product, because they would be rejected by the membranes, along with the desired product, and not separable therefrom by the present process.

The membranes used in the practice of the present process, and generally referred to as reverse osmosis/ultrafiltration membranes, have a nominal pore diameter of 5-15 Angstroms, a preferred range being from 7-11 Angstroms. Membranes useful in the practice of the present invention are manufactured by Osmonics Inc. of Minnetonka, Minn. or by the Celanese Corporation and are generally formulated of cellulose acetate, polyamide or polyvinylfluoride. The filtration is carried out under a hydrostatic pressure of approximately 200 to 400 p.s.i.g. applied to the upstream side of the membrane. By use of a membrane having the appropriate critical pore size, those impurities of a molecular size smaller than the nominal pore diameter of the membrane, along with a large quantity of water, are thus forced through the membrane and accumulate on the downstream side as the permeate, while the desired product molecules, as well as impurities of a molecular size larger than the nominal pore diameter of the membrane, are rejected by the membrane and remain on the upstream side thereof where the product becomes more and more concentrated as more and more water and impurities are forced to the downstream side.

As indicated above, although the membranes used in the present process are referred to generically as reverse osmosis/ultrafiltration membranes, the term "reverse osmosis" generally refers to membranes which reject all solute particles, including ions, and will pass only water molecules, while the term "ultrafiltration" generally refers to membranes which will reject only solute particles above a certain molecular size and will pass smaller particles. (See, for example, Lacey, *Chemical Engineering*, Sept. 4, 1982, page 5). In the context of the present invention, therefore, the term "ultrafiltration" is considered more

appropriate than the term "reverse osmosis" and accordingly is used to describe the invention.

In the preferred practice of the present process, the reaction mixture resulting from the last step in the synthetic procedure, i.e. the diazonium coupling reaction in the preparation of FD and C Red 40, FD and C Yellow 6, FD and C Red 2 or Carmoisine or the sulfonation reaction in the preparation of D and C Yellow 10, is passed to a holding tank and optionally filtered, to remove any insoluble material, before being fed to an ultrafiltration unit where, under a pressure of approximately 200 to 400 p.s.i.g., supplied by a high pressure centrifugal pump, the impurities are forced through the membrane into the permeate which can be collected for analysis or passed directly to waste lines for disposal. Alternatively the crude product previously isolated, by salting out or spray drying, can be redissolved in water and the resulting solution treated as just described.

In one embodiment contemplated by the invention, the solution from the holding tank is fed continuously to the ultrafiltration unit, while solution from the upstream side of the membrane is recirculated back to the tank. Thus ultrafiltration is carried out continuously, the

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concentrate in the tank being continually depleted of impurities, and water is added to the concentrate (to replace that removed in the permeate with the impurities) at such a rate as to maintain the concentration of the product in the concentrate at approximately 5% (w/w). This procedure is referred to hereinafter as diafiltration.

In another embodiment contemplated by the invention, the concentrate from the upstream side of the membrane is recirculated back to the holding tank to be replaced by additional solution fed from the tank. However, instead of replacing the water lost from the system into the permeate as in the diafiltration method, the product is al-

lowed to concentrate in the holding tank, the extent of such concentration, of course, not being allowed to proceed to the point where crystallization of the product would occur. In such instance, additional water is added to insure complete solution of the product at all times so as to obviate plugging of the membrane pores by the crystalline material. Typically the concentration of the product in the concentrate is maintained between approximately 5% and 25% (w/w).

In both of the above-described embodiments, the progressive removal of impurities is followed by sampling the permeate from time to time, and filtration is terminated when essentially no further impurities can be detected in the permeate and, in the case of FD and C Red 40, FD and C Yellow 6, FD and C Red 2 and Carmoisine, when, in addition, the level of sodium naphtholsulfonates, in the concentrate is determined (by appropriate analytical methods such as HPLC, TLC, etc.) to be less than 0.3% of the pure color content. The essential absence of impurities in the permeate can be determined in a variety of ways, such as by determining its electrical conductance. In that method, the conductance of the permeate gradually drops during ultrafiltration because of the continuous removal of ionic species from the concentrate. When the conductance of the permeate drops from an initial level of approximately 50,000 micromhos to approximately 1,000 micromhos, and when the level of sodium naphtholsulfonates in the concentrate reaches the desired level, as indicated above, the removal of essentially all impurities can be considered complete. When that point is reached, the concentrate, containing the highly purified product in water, is evaporated to dryness by any of a number of conventional means, for example, by pan drying or spray drying, in order to isolate the product. In this manner, one can obtain molar yields up to 98% in the process. In contrast, for example, yields of only around 77% of the total available pure color (for FD and C Yellow 6) and 86% of the total available

pure color (for FD and C Red 40) are obtainable using the salting out method of isolation.

In carrying out the present process the reaction mixture, as produced in the diazo coupling and as fed to the ultrafiltration unit, generally has a pH of approximately 9.0. While these solutions can be subjected successfully to ultrafiltration, it is preferred to adjust the pH to approximately 6.0 to 8.0 before passage through the ultrafiltration membrane.

The ultrafiltration process is preferably carried out at ambient temperature but can be carried out at temperatures up to around 40° C.

In order to further describe the invention and the unique advantages afforded thereby, the following examples are included by way of illustration in order to contrast the preparation and purification of FD and C

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Red 40 and FD and C Yellow 6, in accordance with the present invention, with the preparation and purification of the same by conventional methods. The preparation and purification of FD and C Red 2, Carmoisine and D and C Yellow 10, in accordance with the process of the invention further illustrate the same.

EXAMPLE 1

Preparation and Purification of FD and C Red 40
by the Method of the Invention

Diazotization

To a rubber lined 1,500 gallon tank was added 3,550 pounds of water and 1,085 pounds of 5-methoxy-2-methyl-sulfanilic acid, and the pH of the solution was adjusted to 6.0 to 8.0 by the addition of about 550 pounds of 50% sodium hydroxide. The mixture was stirred, and when all material had dissolved, the solution was treated

with 350 pounds of sodium nitrite, stirring until all material had dissolved, and was then cooled to 25°-30° C. by the addition of ice.

To a separate rubber lined 3,000 gallon tank was added 2,300 pounds of water, followed by 1,510 pounds of 20° Bé hydrochloric acid and 2,000 pounds of ice, and the solution was cooled to -5° C. to 0° C. The solution from the 1,500 gallon tank was then pumped slowly into the 3,000 gallon tank while checking the pH frequently in order to maintain acid conditions (blue to Congo Red) and checking frequently for excess nitrite with starch/iodide paper in order to insure that excess nitrite is present during the diazotization. (When all the solution from the first tank has been added, the test for nitrite should be positive, and if necessary an additional 1 to 2 pounds of sodium nitrite is added to give a positive test for nitrite.) The reaction mixture was stirred at 0°-5° C. for about one to one and a quarter hours, while maintaining a slight excess of nitrite ion.

Preparation of SS Solution

To another rubber lined 6,500 gallon tank were added 8,000 pounds of water, 1,000 pounds of sodium carbonate and 1,255 pounds of sodium 2-naphthol-6-sulfonate. The resulting slurry was agitated until uniform and saved for coupling.

Coupling Reaction

The diazotized solution from the 3,000 gallon tank, at 0°-5° C., was then slowly pumped into the 6,500 gallon tank at 20°-25° C. over a one half to one hour period, while testing frequently for excess diazo compound against alkaline H-Acid solution (8-amino-1-naphthol-3,6-disulfonic acid), and if excess diazo compound was detected, the rate of addition of the diazo solution was adjusted to give a continuous negative test. The solution was also tested from time to time to insure a continued excess of sodium 2-naphthol-6-sulfonate against Diazo Blue B solution (2,2-dimethyl-4,4'-bis diazo-biphenyl dichlo-

ride), and to insure that the pH of the solution remains alkaline. (When all the diazo solution has been added, the temperature should be 20°-25° C., the test for diazo compound should be negative, the test for sodium 2-naphthol-6-sulfonate should be positive, and the pH should be 8.3 to 8.8.)

The solution was then stirred for an additional half hour, the pH was adjusted to 6.5 to 6.7 by the addition of 20° Bé hydrochloric acid and treated with 50 pounds of DICALITE® brand of diatomaceous earth and 180 pounds of DARCO® S51 brand of decolorizing char-

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coal. The solution was then heated and stirred at 70°-75° C. for a half hour and then filtered. The filter was washed with about 4,500 pounds of water, and the combined filtrate was adjusted to pH 6.0 to 8.0 by addition of hydrochloric acid and was then led to a holding tank. From that solution was taken a 12 gallon aliquot amounting to 0.2% of the total product, together with impurities which was fed through a high pressure centrifugal pump to an ultrafiltration unit equipped with a cellulose acetate membrane having a nominal pore diameter of 11 Angstroms and under a hydrostatic pressure of 200-400 p.s.i.g. and subjected to diafiltration. That is, the concentrate was recycled back to the holding tank where the concentration of the product was maintained at around 5% by the addition of water. The permeate was collected separately and tested from time to time for its conductance, and the concentrate was tested from time to time for the total amount of SS relative to the total color. After a total of five cycles (of the product solution to the ultrafiltration unit and back to the holding tank), when the conductance had dropped to around 1,000 micromhos, and the amount of SS in the concentrate was less than 0.3% of the pure color content, ultrafiltration was interrupted. During the filtration the total pure color that passed through the membrane was determined, by either spectrometric meth-

ods or by visual comparison with known color standards, to constitute about 2% of the total available color in the original unfiltered solution from the reaction mixture thus leaving 98% of the total available pure color in the concentrate. The ultrafiltration process as described above afforded 2 gallons of purified concentrate. From this concentrate was take a further 600 ml. aliquot which was spray dried to give 150 g. of purified product which, on assay, had the following specifications, the range of specifications required by regulations of the Food and Drug Administration being included for purposes of comparison. Here, and in all tables which follow, unless noted otherwise, all values are given in percent.

	Found	FDA Spec.
Pure Color	91.9	85
NaCl	0.03	14*
Na ₂ SO ₄	0.56	
Volatiles	6.14	
CSA	<0.02	0.2
SS	0.01	0.3
DMMA	<0.02	0.1
HS	<0.3	1.0
DONS	<0.05	1.0

* The FDA specifications require that the total amount of NaCl, Na₂SO₄, and Volatiles be not more than 14%. In each of the assays reported herein, separate values for each of these entries were determined and are recorded. The totals, in each case, will be seen to be within the required limits.

Two further samples of FD and C Red 40, prepared as described above, were purified by diafiltration using the procedure described above except that in one run a polyamide membrane having a nominal pore diameter of 7-10 Angstroms (Zero PA membrane obtained from the Celanese Corporation) was used and in a second run a polyvinylfluoride membrane having a nominal pore diameter of 10 Angstroms (20 VF membrane from Os-

monics, Inc.) was used, to give 97% recovery of product in each case. The samples so purified had the following specifications, the ranges of specifications required by FDA regulations being given again for comparative purposes.

[10]

	Found		FDA Spec.
	Zero PA	20 VF	
Pure Color	88.4	89.3	85
NaCl	0.05	0.06	14
Na ₂ SO ₄	0.78	0.67	
Volatiles	9.22	9.41	
CSA	<0.02	<0.02	0.2
SS	0.03	<0.02	0.3
DMMA	0.16	<0.02	0.1
HS	0.3	0.3	1.0
DONS	<0.05	<0.05	1.0

Preparation and Purification of FD and C Red 40 by the Prior Method

The above procedure was repeated through the filtration of the solution from the coupling reaction and the washing of the filter with 4,500 pounds of water. The combined filtrate was transferred to a 7,000 gallon stainless steel crystallization tank. To the tank was added 9,000 pounds of salt (equivalent to 18% of the solution volume) over a period of one half to one hour and while maintaining the temperature at about 65° C.

The crystalline material which separated was collected by filtration, and the solid was washed on the filter sequentially with 1,200 gallons each of 18° Be and 12° Be brine at 0° C. to 5° C. The filter was given a final wash with a solution of 750 gallons of water and 450 gallons of ethyl alcohol, and the product was collected

and dried. There was thus obtained 2,170 pounds (87.5% yield based on 5-methoxy-2-methylsulfanilic acid) of the disodium salt of 1-[(6-methoxy-4-sulfo-3-methylphenyl)azo]-2-naphthol-6-sulfonic acid.

The material so-obtained in a series of similar runs was assayed in each case, in accordance with Food and Drug Administration regulations, and found to have the following ranges of specifications, the specifications obtained with material purified in accordance with the process of the invention as described above and specifications required by regulations of the Food and Drug Administration being given for purposes of comparison.

	Found (%)	Claimed Process	FDA Spec. (%)
Pure Color	88-92	91.9	85
NaCl	2.0-3.5	0.03	14
Na ₂ SO ₄	0.05-0.1	0.56	
Volatiles	3.3-7.0	6.14	
CSA	0.02	<0.02	0.2
SS	0.02-0.2	0.01	0.3
DMMA	0.02	<0.02	0.1
HS	0.2-1.0	<0.05	1.0
DONS	0.1-0.2	<0.05	1.0

EXAMPLE 2

Preparation and Purification of FD and C Yellow 6 by the Method of the Invention

Diazotization

To a rubber lined 1,500 gallon tank was added 2,000 pounds of water followed by 1,038 pounds of sulfanilic acid and 490 pounds of sodium hydroxide, and the mixture was heated to 45° C. and stirred until all material dissolved. Additional sodium hydroxide was added as

necessary to make the solution alkaline to Brilliant Yellow.

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To the resulting solution was added, slowly and with stirring, 1,800 pounds of 20° Bé hydrochloric acid. When addition was complete, the mixture, which consisted of a slurry of sulfanilic acid in the liquid phase, was tested for acidity to Congo Red, and additional hydrochloric acid added as necessary to adjust the pH accordingly. The mixture was then cooled to 0° C. by addition of ice (about 3,000 pounds), and the solution was treated slowly, over a five to ten minute period, with a solution of 420 pounds of sodium nitrite in 1,000 pounds of water, while maintaining the temperature at 10°-12° C., the solution being added at such rate that no nitrous oxide was given off from the mixture. (When addition of the sodium nitrite is complete, the mixture should be positive to nitrite, and if not an additional 1 to 2 pounds of sodium nitrite are added to insure a slight excess.)

Preparation of SS Solution

In a separate rubber lined 6,500 gallon tank containing 5,000 pounds of water was added 1,480 pounds of sodium 2-naphthol-6-sulfonate, and the mixture was stirred until a smooth slurry was obtained. The pH was adjusted to 9.3 to 9.5 with 50% sodium hydroxide and then cooled, if necessary, to 20°-25° C. with ice.

Coupling Reaction

The diazo solution from the first tank, at 0°-5° C., was then pumped into the second tank over about a half hour period while maintaining the pH at 8.5 to 9.0 by addition of 50% sodium hydroxide, testing frequently for excess diazo compound with alkaline H-Acid. If excess diazo compound was detected, the rate of addition of the diazo solution was adjusted to give a continuous negative test. The solution was also tested from time

to time for excess sodium 2-naphthol-6-sulfonic acid against Diazo Blue B solution in order to insure the continuous presence of an excess thereof. (When all the diazo solution has been added, the temperature should be 20°-25° C., the test for excess diazo compound should be negative, the test for sodium 2-naphthol-6-sulfonate should be positive, and the pH should be 8.4 to 9.0.)

The solution was then stirred for an additional half hour, the pH was adjusted to 6.5 to 6.7 by the addition of 20° Bé hydrochloric acid and treated with 50 pounds of DICALITE® and 180 pounds of DARCO® S51. The solution was then heated and stirred at 70°-75° C. for a half hour and filtered. The filter was washed with about 4,500 pounds of water, and the combined filtrate was adjusted to pH 6.0 to 8.0 by addition of hydrochloric acid and was then led to a holding tank. From that solution was taken a 9 gallon aliquot, amount to 0.2% of the total product together with impurities, which was then fed through a high pressure centrifugal pump to an ultrafiltration unit equipped with a cellulose acetate membrane having a nominal pore diameter of 11 Angstroms and under a hydrostatic pressure of 200-400 p.s.i.g., the concentrate being recycled back to the holding tank where the concentration of the product was maintained at approximately 5% by addition of water. The permeate was collected separately and tested from time to time for its conductance, and the concentrate was tested from time to time for the total amount of SS relative to the total color. After a total of five cycles (of the product solution to the ultrafiltration unit and back to the holding tank), when the conductance of the permeate had dropped to around 1,000 micromhos, and the

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amount of SS in the concentrate was less than 0.3% of the pure color content, diafiltration was interrupted. During the filtration the total pure color that passed through

the membrane was determined, by either spectrometric methods or by visual comparison with known color standards, to constitute approximately 5% of the total available color in the original unfiltered solution from the reaction mixture thus leaving 95% of the total available pure color in the concentrate. The ultrafiltration process as described above afforded 4.5 gallons of purified concentrate. From this concentrate was taken a further 800 ml. aliquot which was spray dried to give 54 g. of purified product which, on assay, had the following specifications, the range of specifications required by regulations of the Food and Drug Administration being included for purposes of comparison.

	Found	FDA Spec.
Pure Color	92.2	85
NaCl	0.26	14
Na ₂ SO ₄	2.80	
Volatiles	4.89	
SA	0.02	0.2
SS	0.1	0.3
DAAB	0.02	0.1
DONS	<0.2	1.0

Preparation and Purification of FD and C Yellow 6 by the Prior Method

The above procedure was repeated through the filtration of the solution from the coupling reaction and the washing of the filter with 4,500 pounds of water. The combined filtrate was transferred to a 7,000 gallon stainless steel crystallization tank. To the tank was added, over a period of a half hour at 70° C., an amount of sodium chloride equivalent to about 17% of the total volume (8,000-9,000 pounds).

The crystalline material which separated was collected by filtration, and the solid was washed on the filter with 1,200 gallons of 18° Bé brine, then four times with 2°

Bé brine at 0°-2° C. (1,200 gallons per wash), three more times with 18° Bé brine (1,200 gallons per wash) and finally two times with 1,200 gallons of water at 0° C. and then dried. There was thus obtained 2,100 pounds of the disodium salt of 1-[(4-sulphophenyl)azo]-2-naphthol-6-sulfonic acid (77% yield based on sodium 2-naphthol-6-sulfonate).

The material so-obtained in a series of similar runs was assayed in each case, in accordance with Food and Drug Administration regulations, and found to have the following range of specifications, the specifications obtained with material purified in accordance with the process of the invention as described above and specifications required by regulations of the Food and Drug Administration being given also for purposes of comparison.

	Found	Claimed Process	FDA Spec.
Pure Color	89-92	92.2	85
NaCl	3.4-5.0	0.26	14
Na ₂ SO ₄	<0.05	2.80	
Volatiles	1.7-5.4	4.89	
SA	0.02	0.02	0.2
SS	0.04-0.08	0.1	0.3
DAAB	0.02	0.02	0.1
[13]			
DONS	0.07-0.2	<0.02	1.0

EXAMPLE 3

Preparation and Purification of FD and C Red 2

Diazotization

A 10 liter glass reactor was charged with 7.5 liters of tap water and 965 g. of sodium 4-amino-1-naphthalene sulfonate (76%, 3 moles). The mixture was stirred until

the sulfonate dissolved, and the solution was then treated with 30 g. of NORIT® FQA brand of decolorizing charcoal and the resulting slurry filtered. The filtrate was acidified with 911 g. of 20° Bé hydrochloric acid, and the resulting slurry was cooled with ice to 5°-10° C. and diazotized by the dropwise addition of 500 ml. of an aqueous solution of 209 g. (3.03 moles) of sodium nitrite over a one and one quarter hour period, while maintaining the temperature and pH throughout the addition at <10° C. and 1, respectively. When all the nitrite had been added, the diazonium salt slurry was stirred at 0°-10° C. and pH 1 for three hours, and the presence of excess nitrous acid was verified periodically by testing with starch/potassium iodide paper, additional sodium nitrite being added to maintain a positive test.

Coupling Reaction

A 20 liter glass reactor was charged with 6.3 liters of tap water, 1328 g. of disodium 2naphthol-3,6-disulfonate (81%, 3.09 moles) and 569 g. (5.37 moles) of sodium carbonate. The mixture was stirred until the sodium carbonate and the disulfonate salt had dissolved, and the diazonium salt slurry from the previous step was added to the solution over a ninety minute period, while maintaining the temperature and the pH at 18°-25° C. and 8-10, respectively, and while testing frequently for excess diazo compound with alkaline H-Acid in order to insure a continuous negative test with respect to the diazo compound. The solution was then treated with 7 g. of NORIT®FQA brand of activated charcoal and 35 g. of DICALITE® brand of diatomaceous earth and then heated to 55° C. for two and a half hours. The slurry was then filtered, cooled to room temperature and the filtrate subjected to diafiltration as described above using a cellulose acetate membrane having a nominal pore diameter of 11 Angstroms until the conductance of the concentrate and the permeate levelled off at 11,000 micromhos and 650 micromhos, respectively. The concentrate was then fur-

ther concentrated to about 3.75 gallons by ultrafiltration. A 500 ml. aliquot of this concentrate was spray dried to give 67 g. of FD and C Red 2 (Amaranth) powder, corresponding to a total pure color recovery of 1772 g. or 98% of theory. This material, on assay, had the following specifications, the specifications required by EEC regulations being included for purposes of comparison.

	Found	EEC Spec.
Pure Color	91.0	85
NaCl, Na ₂ SO ₄	1.04	5
Volatiles	7.04	10
[14]		
Subsidiary Colors	0.9	3

EXAMPLE 4

Preparation and Purification of Carmoisine by the Method of the Invention

Diazotization

A 10 liter glass reactor was charged with 5.5 liters of water and 772 g. (76%, 2.24 moles) of sodium 4-amino-1-naphthalene sulfonate, and the mixture was stirred until the sulfonate dissolved. The resulting solution was treated with 30 g. of NORIT® FQA brand of activated charcoal, the slurry was filtered, and the filtrate was acidified with 878 g. of 20° Bé hydrochloric acid. The resulting slurry was then cooled to 5°-10° C. and diazotized by the dropwise addition of 500 ml. of an aqueous solution of 156 g. (2.26 moles) of sodium nitrite over a one and one quarter hour period, while maintaining the temperature and pH of the reaction mixture at 10° C. and 1, respectively. When all the nitrite solution had been added, the resulting slurry was stirred at 0°-10° C. and pH of about 1 for three hours while testing periodically with starch/

potassium iodide paper to insure a slight excess of nitrous acid at all times.

Coupling Reaction

A 20 liter glass reactor was charged with 5.5 liters of water and 425 g. of sodium carbonate, the solution was stirred until the carbonate dissolved, and then 630 g. (91.7%, 2.35 moles) of sodium 1-naphthol-4-sulfonic acid was added with stirring until all material had dissolved. The solution was then cooled to 5° C., and the previously prepared solution of the diazonium salt was then added over a period of approximately one hour while maintaining the temperature and pH at 5°-6° C. and 9-11, respectively.

The pH of the resulting solution was adjusted to 6-7 and subjected to diafiltration through a cellulosic acetate membrane having a nominal pore diameter of 11 Angstroms at 200-400 p.s.i.g. and 2-3 gallons per minute. The diafiltration was continued until the concentrate and the permeate conductivities had leveled off at 5,000-10,000 micromhos and <1,000 micromhos, respectively, and the concentrate was then further concentrated to a total volume of about 4 gallons. A 500 ml. aliquot of this concentrate was spray dried to give 41 g. of pure dye, corresponding to a total recovery of 98% of theory. The material, on assay, had the following specifications, the specifications required by regulations of the E.E.C. being provided for purposes of comparison.

	Found	EEC Spec.
Pure Color	89.0	85
NaCl	0.07	N/A
Na ₂ SO ₄	2.26	N/A
Volatiles	1.64	N/A
Subsidiary Colors	<1.0	1.0
Unreacted Intermediates	<0.5	0.5

[15]

EXAMPLE 5

Purification of D and C Yellow 10 by the Method of the Invention

To a 22 liter glass reactor charged with 16 liters of distilled water was added 803 g. of Quinoline Yellow WS (approximately 60% pure dye), and the solution was stirred until the solid had dissolved. The solution was then treated with 288 g. of DARCO® S-51 brand of activated charcoal and 100 g. of DICALITE® brand of diatomaceous earth, and the resulting mixture was heated to 80°-90° C. for two hours and then filtered. The filtrate was cooled to 30°-40° C. and then subjected to diafiltration through a Zero PA polyamide membrane having a nominal pore diameter of 7-10 Angstroms until the conductance of the permeate and the concentrate levelled off at 200-500 micromhos and 5,000-7,000 micromhos, respectively. The concentrate was then further concentrated to 3.5 gallons and a 1 gallon aliquot thereof was spray dried to give 103 g. of purified material, corresponding to 75% total recovery. The purified sample thus obtained, on assay, gave the following specifications, the corresponding specifications of the crude material prior to ultrafiltration and the specifications required by FDA regulations being provided for purposes of comparison.

	Found		FDA Spec.
	Crude	Purified	
Pure Color	60	89	85
NaCl	12	0.1	15
Na ₂ SO ₄	20	4.0	
Volatiles	8.0	6.1	
Yellow 11	108 ppm	<0.1 ppm	4 ppm
Chlorinated Yellow 11	1190 ppm	<0.1 ppm	2 ppm

We claim:

1. In a process for the purification of a dye selected from the group consisting of the disodium salt of 1-[(6-methoxy-4-sulfo-3-methylphenyl)azo]-2-naphthol-6-sulfonic acid, the disodium salt of 1-[(4-sulfophenyl)azo]-2-naphthol-6-sulfonic acid, the trisodium salt of 1-[1-(4-sulfonaphthyl)azo]-2-naphthol-3,6-disulfonic acid, the disodium salt of 2-[1-(4-sulfonaphthyl)azo]-1-naphthol-4-sulfonic acid and the sodium salt of 2-(2-quinolyl)-1,3-indanedione-sulfonic acid as the products resulting, respectively, from the diazotization of 5-methoxy-2-methylsulfanilic acid in water with sodium nitrite in the presence of hydrochloric acid followed by the coupling under alkaline conditions of the resulting 5-methoxy-4-sulfo-2-methylphenyldiazonium chloride with sodium 2-naphthol-6-sulfonate; the diazotization of sulfanilic acid in water with sodium nitrite in the presence of hydrochloric acid followed by the coupling under alkaline conditions of the resulting 4-sulfophenyldiazonium chloride with sodium 2-naphthol-6-sulfonate; the diazotization of 4-aminonaphthalene-1-sulfonic acid in water with sodium nitrite in the presence of hydrochloric acid followed by the coupling under alkaline conditions of the resulting 1-sulfonaphthyl-4-diazonium chloride with disodium 2-naphthol-3,6-disulfonate; the diazotization of 4-aminonaphthalene-1-sulfonic acid in water with sodium nitrite in the presence of hydrochloric acid followed by the coupling under alkaline conditions of the resulting 1-sulfonaphthyl-4-diazonium chloride with sodium 1-naphthol-4-sulfonate; and the condensation of 2-quinaldine with phthalic anhydride followed by sulfo-

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nation of the resulting 2-(2-quinolyl)-1,3-indanedione, said dye being present in the resulting reaction mixtures, along with impurities, the improvement which comprises: subjecting an aqueous solution of the reaction mixture resulting from said coupling or said sulfonation to ultrafiltration through a membrane having a nominal

pore diameter of 5-15 Angstroms under a hydrostatic pressure of approximately 200 to 400 p.s.i.g., at a pH from approximately 6.0 to 9.0, to thereby cause separation of said impurities from said dye, said impurities of a molecular size smaller than the nominal pore diameter passing into the permeate on the downstream side of said membrane and said dye remaining in the concentrate, and when substantially all said impurities have been removed from said concentrate, as evidenced by their essential absence in said permeate, recovering said dye, in approximately 90% purity from said concentrate by evaporation of said concentrate to dryness.

2. A process according to claim 1 wherein said membrane is composed of cellulose acetate, polyamide or polyvinylfluoride.

3. A process according to claim 2 wherein the concentration of the dye in said concentrate is maintained at approximately 5-25% (w/w) by recycling the concentrate and adding water thereto.

4. A process according to claim 2 wherein ultrafiltration is carried out until the concentration of the azo dye in the concentrate is maintained at approximately 5% (w/w).

5. A process according to claim 3 wherein the pH of the reaction mixture is adjusted to 6.0 to 8.0 before ultrafiltration.

6. A process according to claim 4 wherein the pH of the reaction mixture is adjusted to 6.0 to 8.0 before ultrafiltration.

7. A process according to claim 5 wherein ultrafiltration is interrupted, and the product isolated, when the conductance of the permeate is approximately 1,000 micromhos.

8. A process according to claim 6 wherein ultrafiltration is interrupted, and the product isolated, when the

conductance of the permeate is approximately 1,000 micromhos.

9. A process according to claim 7 wherein the product is isolated by spray drying of the concentrate.

10. A process according to claim 7 wherein the product is isolated by pan drying of the concentrate.

11. A process according to claim 8 wherein the product is isolated by spray drying of the concentrate.

12. A process according to claim 8 wherein the product is isolated by pan drying of the concentrate.

13. A process according to claim 1 wherein said dye is the disodium salt of 1-[(6-methoxy-4-sulfo-3-methylphenyl)azo]-2-naphthol-6-sulfonic acid.

14. A process according to claim 1 wherein said dye is the disodium salt of 1-[(4-sulfophenyl)azo]-2-naphthol-6-sulfonic acid.

15. A process according to claim 1 wherein said dye is the trisodium salt of 1-[1-(4-sulfonaphthyl)azo]-2-naphthol-3,6-disulfonic acid.

16. A process according to claim 1 wherein said dye is the disodium salt of 2-[1-(4-sulfonaphthyl)azo]-1-naphthol-4-sulfonic acid.

17. A process according to claim 1 wherein said dye is the sodium salt of 2-(2-quinolyl)-1,3-indanedione-sulfonic acid.

* * * *

[Filed Apr. 23, 1991]

IN THE UNITED STATES DISTRICT COURT
FOR THE SOUTHERN DISTRICT OF OHIO
WESTERN DIVISION

Civil Action No. C-1-91-218

(Weber, J.)

HILTON DAVIS CHEMICAL CO.,
Plaintiff,

v.

WARNER-JENKINSON,
Defendant.

ANSWER

Defendant, through its attorneys, hereby answers the Complaint herein as follows:

1. Defendant admits that plaintiff's First Cause of Action purports to state a cause of action for patent infringement under Title 35, United States Code, but denies that any such cause of action exists. Defendant admits that this Court has jurisdiction of this cause of action and that venue is proper in this district.

2. On information and belief, defendant admits the allegation of paragraph 2.

3. Defendant admits the allegation of paragraph 3.

4. Defendant admits that a copy of U.S. Patent No. 4,560,746 was attached to the Complaint as Exhibit A and shows on its face that it was issued on December 24, 1985, for an Ultrafiltration Process for Purification of Dyes Useful in Foodstuffs, and was assigned to the Hilton-Davis Chemical Co. Defendant denies, however, that said patent was duly and legally issued.

5. Defendant denies the allegation of paragraph 5.

Affirmative Defenses

1. Defendant has not infringed U.S. Patent No. 4,560,746.

2. Upon information and belief, U.S. Patent No. 4,560,746 is invalid for failure to comply with the statutory requirements of Title 35 of the United States Code.

WHEREFORE, defendant demands as follows:

A. That judgment be entered in its favor on the Complaint and that the Complaint be dismissed with prejudice and plaintiff take nothing by its action.

B. That defendant be awarded its costs and attorney fees incurred in the defense of this action.

C. That defendant have such other and further relief as this Court may deemed just and proper.

WARNER-JENKINSON COMPANY, INC.

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JURY INSTRUCTIONS

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[2028] THE COURT: Ladies and gentlemen, I'm going to ask [2029] Mrs. Jones to distribute a copy of the charge or the instructions of law to you. You can follow along or not, just however you choose. It is my duty to read them into the record at this time. The instructions that I'm reading from are here and we will put them in the notebook for your information with the verdict forms.

Members of the jury, your job as jurors is to determine the issues of fact presented by the claims of the parties and reach a just verdict.

In doing your job, you must follow the law as stated in these instructions and apply these rules of law to the facts you find from the legal evidence.

You are not to single out one instruction alone as stating the law, but must consider the instructions as a whole.

You are not to be concerned with the wisdom of any rule of law. Regardless of any opinion you may have as to what the law ought to be, it would be a violation of your sworn duty to base a verdict upon any other view of the law than that given in these instructions, just as it would be a violation of your sworn duty as judges of the facts to base a verdict upon anything but the legal evidence in the case.

You are to perform this duty without bias or prejudice as to any party. Our system of law does not permit jurors to be governed by sympathy, prejudice, or public opinion. Both the parties and the public expect that you will [2030] carefully and impartially consider all the legal evidence in the case, follow the law as stated by the

Court, and reach a just verdict regardless of the consequences.

This case should be considered and decided by you as an action between persons of equal worth and equal standing in the community. Businesses, corporations and governmental units are entitled to the same fair trial at your hands as private individuals. All persons, including businesses, corporations and governmental units, stand equal before the law and are to be dealt with as equals in a court of justice. Sympathy for a party or prejudice against a party should play no part in your deliberations or in your decision.

Unless otherwise stated, the jury should consider each instruction given to apply separately and individually to each party.

The person who asserts a claim has the burden of proof, that is, he or she has the obligation to prove it by a certain level of proof.

There are two levels of proof involved in this case: proof by clear and convincing legal evidence and proof by a preponderance of the legal evidence.

The defendant has the burden of establishing the invalidity of the claims of the patent by clear and convincing evidence. The plaintiff has the burden of proving infringement and damages, if any, by a preponderance of the legal evidence.

[2031] The plaintiff has the burden of establishing that the defendant's infringement of the claims of the patent by a defendant, if any, was willful by clear and convincing evidence.

It is the quality of the legal evidence that must be weighed. Quality is not necessarily identical with quantity or the greater number of witnesses.

In determining whether any fact in issue has been proved by the legal evidence in the case, you may, unless otherwise instructed, consider the testimony of all wit-

nesses, regardless of who may have called them, and all exhibits received as legal evidence, regardless of who may have produced them.

Preponderance of the legal evidence is the greater weight of the evidence. That is, legal evidence that you believe because it outweighs or overbalances in your minds the legal evidence opposed to it and because it is more probable, more persuasive or of greater probative value.

If the weight of the legal evidence is equally balanced or if you are unable to determine which side of an issue has been proved by a preponderance of the legal evidence, the party who has the burden of proof has not established such issue by a preponderance of the legal evidence.

This rule of law does not, of course, require proof to an absolute certainty, since proof to an absolute certainty [2032] is seldom possible in any case.

It is proper to find that a party has succeeded in carrying the burden of proof by a preponderance of the legal evidence on an issue of fact, if, after consideration of the legal evidence, the jurors believe that what is sought to be proved on that issue is more likely true than not true.

Clear and convincing legal evidence is legal evidence which produces in your mind a firm belief that the truth of the factual contentions are highly probable. Clear and convincing proof is proof of the facts which lies somewhere between preponderance of the evidence and beyond a reasonable doubt.

Legal evidence is the testimony of witnesses, the exhibits which you will have with you in the jury room, regardless of who may have produced them, and the facts which have been admitted or stipulated.

Once facts have been proved by the legal evidence, you may then draw such reasonable inferences from those facts you feel are justified in the light of your experience.

The legal evidence does not include the exhibits which have not been given to you in the jury room, the opening statements or the closing arguments of counsel. The opening statements and closing arguments of counsel are designed to assist you. They are not legal evidence.

Statements or answers which were stricken by the [2033] Court or which you were instructed to disregard are not legal evidence and must be treated as though you never heard them. Likewise, anything you may have perceived outside the courtroom is not legal evidence and must be entirely disregarded.

You must not speculate as to why the Court sustained the objection to any question or what the answer to such question might have been. You must not draw any inference or speculate on the truth of any suggestion included in a question that was not answered. It is the duty of the attorneys on both sides to make their objections and protect the interests of their clients. You are not to infer in any manner that either side was attempting to withhold any evidence from you as a result of objecting to the evidence, whether it was subsequently admitted by the Court or subsequently ruled inadmissible.

You must not draw any inference or speculate on the truth of any suggestion included in a question that was not answered.

If a lawyer asks the witness a question which contains an assertion of fact, you may not consider the assertion of fact as legal evidence of that fact unless adopted or confirmed by the witness. The lawyer's assertion of facts are not legal evidence.

The questions of lawyers may be considered, however, to the extent that they give meaning to the answers of the [2034] witnesses.

There are, generally speaking, two types of legal evidence which the jury may properly find the truth as to the facts of the case.

Direct evidence is testimony given by a witness who has personal knowledge of the facts to which he or she testifies.

Circumstantial evidence is the proof of facts or circumstances by direct evidence from which you may reasonably infer other related or connected facts which naturally and logically follow, according to the common experience of people.

To infer, or to make an inference, is to reach a reasonable conclusion of fact which you may, but are not required to, make from other facts which you find have been established by direct evidence. Whether an inference is made rests entirely with you.

You may not build one inference from another inference which is unsupported by any additional facts, but you may make more than one inference from the same facts or circumstances if it is reasonable to do so. Additionally, you may not draw inferences from a speculative or remote basis that has not been established by the legal evidence.

As a general rule, the law makes no distinction between direct and circumstantial evidence, but simply requires that you find the facts in accordance with the legal evidence [2035] in the case, both direct and circumstantial.

In your effort to determine the facts, you will be faced with the problem of what weight should be given to the testimony of each witness. You must determine how credible or believable any witness is. You may believe all that a witness tells you, part of what he or she tells you, or none of what he or she tells you.

Consider carefully the circumstances under which each witness testified. Remember the response to questions, assurance or lack of it in answering, and the entire demeanor or appearance of the witness while on the wit-

ness stand. Consider also any relationship that may bear to either side of the case, their reasons for testifying, or any interest they may have in the outcome of the case, any prejudice or bias they may have shown, including any reason or motivation to bear hostility or animosity toward any party, and any partiality they may have demonstrated.

In dealing with contradictory testimony, you may consider which testimony is supported or contradicted by the exhibits admitted into legal evidence.

Material inconsistencies or discrepancies in the testimony of a witness or between the testimony of different witnesses may or may not cause you to discredit that testimony. In considering the effect of a discrepancy, do not be misled by unimportant detail, but do consider all discrepancies that [2036] relate to matters of importance. Consider also whether a discrepancy resulted from an innocent error or an intentional falsehood.

Keep in mind that two individuals rarely, if ever, describe an incident precisely alike in all minute details. Note that one individual rarely, if ever, describes the same incident twice in the same minute detail.

In your daily life, you are constantly determining who is worthy of belief and who is not. In this case, employ the same tests in determining who is worthy of belief and who is not. Employ the same tests in determining the weight and credibility, if any, you will assign to the testimony of each witness who testified in this trial.

If you believe that witnesses have been discredited as to a part of their testimony, you may give the balance of their testimony such credence, if any, that you believe it deserves.

During this trial, various witnesses have been asked if consultation with counsel had occurred before the witness' appearance in court. It is not improper for an at-

torney to interview a witness before he or she testifies. It is a customary and accepted procedure. No possible suggestion of impropriety results from an interview alone.

In this case, you have heard testimony of witnesses who have rendered opinions. Persons who by education, training [2037] or experience have become experts in any art, science, profession or calling, may state their opinions in a matter in which they are experts and which is relevant to the case. They may also state the reasons for their opinion.

You should consider each expert opinion received in this case separately and give it such weight as you think it deserves. You may consider the education, training and experience of the expert witnesses when determining the weight of their opinions. You may reject the expert opinion in whole or in part if you conclude that the reasons given in support of the opinions are unsound.

questions have been asked in which expert witnesses were permitted to assume that certain facts were true and to give their opinions based on those assumptions. You must determine whether the assumed facts upon which the experts base their opinions are true. If any assumed fact was not established by the legal evidence, you must determine the effect of that fact not being established upon the value of the opinion of the expert.

As with other witnesses, however, you must decide what weight should be given the testimony of each expert. In determining its weight, you may take into consideration the experts witness' knowledge, experience, education, truthfulness and familiarity with the facts of the case, as well as other means of testing credibility and determining the weight to be [2038] given to the testimony. In short, you may use all those tests you ordinarily use in everyday life to determine whether an individual is well-informed regarding the matters he or she has talked about.

You will recall that during the trial expert witnesses were asked if they had been or will be compensated for their services. It is not improper for an expert witness

to be compensated for services. It is a customary and accepted procedure. No possible suggestion of impropriety results from compensation for the services of an expert witness, including compensation for giving testimony at trial.

A number of exhibits and testimony relating to them have been introduced. You will determine what weight, if any, the exhibits should receive in light of all the legal evidence.

During the trial in this case, certain testimony has been given by way of deposition. The testimony of a witness, who for some reason cannot be present to testify from the witness stand, may be presented in writing or by videotape under oath, in the form of a deposition. The testimony is entitled to the same consideration and is to be judged as to credibility and weighed, and otherwise considered by you in the same way as if the witness had been present, and had testified from the witness stand.

Summaries or charts prepared by a witness or a party and submitted to you in the jury room are received for the [2039] purpose of explaining facts disclosed by testimony and other documents which are legal evidence in the case. Such summaries or charts, however, are not in and of themselves proof of any facts. If such summaries or charts do not correctly reflect the facts or figures shown by the legal evidence in the case, you may disregard them entirely.

At law, a corporation is a person and must be regarded as such.

The acts of corporate officers and employees within the scope of their employment are the acts of that corporation.

Knowledge of corporate business received by an employee in the ordinary course of business is imputed to the corporation.

Article 1, Section 8 of the United States Constitution Congress was given the power to promote the progress of the useful arts by securing for limited times to inventors the exclusive right to their discoveries.

A patent office, now known as the Patent and Trademark Office, sometimes we refer to it as the PTO, and a system of issuing patents has existed ever since. Patents are granted as an incentive to inventors to develop and disclose to the public advancements in the useful arts.

An inventor is rewarded by a seventeen-year patent when he has made a patentable innovation. During the seventeen-year period, he has a right to prevent others from [2040] making, using or selling his invention. To protect those rights, he may invoke the power of the federal courts.

If the PTO acts favorably upon the inventor's application, it will issue a patent. The patent is divided into two parts. The first part being called the specification and the second part being called the claims. The scope or coverage of a patent is limited only to that which is contained in the claims section of the patent.

The claims of the patent define what the invention is and what others are precluded from making, using or selling.

Three elements requisite to an invention are novelty, utility, and non-obviousness.

To be "new" or "novel," as that word is used in the patent law, the invention must be one that was never before disclosed in a single prior process or a single prior patent or a single prior publication.

To have "utility," means an invention must have some usefulness. To be non-obvious, as that word is used in the patent law, the invention as a whole must not have been obvious to a person having ordinary skill in the pertinent

art at the time the invention was made, in view of the whole of the prior art.

Thus, new, non-obvious and useful things which add to the sum of useful knowledge are inventions which are patentable.

[2041] Another fundamental principle of American patent law is that a person is not entitled to a patent if he did not invent the subject matter claimed in the patent, therefore, the patent law provides that a person shall be entitled to a patent unless he did not himself invent the the subject matter sought to be patented. Being an inventor is a preliminary legal requirement for a valid patent.

The threshold question in determining inventorship is who conceived the invention. Unless a person contributes to the conception of the invention, he is not an inventor. An individual must contribute to the final conception of that which is covered by the claims of the subject patent in order to be considered an inventor. He how first brought the process to perfection and made it capable of useful operation is the inventor and is entitled to the patent. One who merely suggests an idea of a result to be accomplished, rather than means of accomplishing it, is not a joint inventor.

An inventor need not himself undertake all the steps necessary to reduce the invention to practice in order to be an inventor. An inventor made use the services, ideas and aid of others in perfecting his invention without losing his right to a patent. The work, experiments, and suggestions of others in carrying out the conception of an inventor, not rising to the level of invention, do not entitle others to be treated as inventors or co-inventors. Also, the sole fact that someone [2042] other than the inventor was the first to observe an effect or useful property of the invention does not make that party an inventor or deprive the actual inventors of their rights.

Joint invention means collaboration of effort to produce a complete and operative invention. If more than one person contributed to what is claimed in the patent, all of these true inventors must apply together for the patent in their own names, and all of their names must appear on the issued patent in order for it to be valid.

The inventors as named in an issued patent are presumed to be correct. Misjoinder or non-joinder of inventors must be proved by clear and convincing legal evidence by the defendant.

The law of patents provides that a person is entitled to a patent unless before the applicant's invention thereof, the invention was made in this country by another who had not abandoned, suppressed or concealed it. To determine priority of invention, you must consider conception, reduction to practice, and corroboration.

Conception is the initial act of invention. It is the formation in the mind of the inventor of a definite and permanent idea of the complete and operative invention, as it is to be applied in practice. Conception must be proved by corroborating evidence which shows that the inventor disclosed to others his completed thought expressed in such clear terms [2043] to enable those skilled in the art to make the invention. The conception must have been of the invention defined in the claims.

Reduction to practice can be either actual reduction to practice or constructive reduction to practice. Actual reduction to practice occurs when the device is embodied in a physical form sufficient to demonstrate that it will work practically for its intended purpose. That is, actual reduction to practice requires that an invention be sufficiently tested to demonstrate that it will work for is intended purpose. There cannot be an actual reduction to practice of the invention without an actual physical embodiment which includes all limitations of the claims. Commercialization of the invention need not be demon-

strated for actual reduction to occur. Constructive reduction occurs on the date when the patent application is filed.

Finally, conception and reduction to practice must be corroborated or substantiated by evidence in addition to the inventor's own statements and documents.

You must consider all the facts of this case to determine whether there has been an abandonment, suppression or concealment. Public use of the invention, without disclosing the details of it, is sufficient to negate any intention to abandon, suppress or conceal. Engaging in activities designed to bring about public or commercial use of the invention is [2044] also sufficient.

Every patent issued by the United States Patent and Trademark Office is presumed to be valid as to each claim independently of the other claims. That presumption, however, is not absolute. You must also presume that a government agency, such as the United States Patent and Trademark Office, has properly done its job in examining the Hilton Davis patent until you are clearly convinced to the contrary.

In order to rebut such presumptions, defendant must establish by clear and convincing legal evidence the invalidity of each claim it asserts to be invalid. That burden of persuasion never switches from the patent challenger—never shifts from the patent challenger on this issue.

U.S. Patent No. 4,560,746 entitled "Ultrafiltration Process for Purification of Dyes Useful in Foodstuffs" issued on December 24 1985. The patent names as inventors: Robert W. Rebhahn and Wayne L. Cook. Application to the patent office for the Hilton Davis patent was originally made on March 28, 1983. A continuation-in-part application was filed on November 30, 1984.

The plaintiff in this action, Hilton Davis, is the owner of the patent. The defendant is Warner-Jenkinson company. Both Hilton Davis and Warner-Jenkinson manufacture FDA certifiable food dyes.

Plaintiff Hilton Davis contends that its patent [2045] number 4,560,746 is valid and leaves the burden of proving invalidity on Warner-Jenkinson. I have asked the parties to refer to the patent as the Hilton Davis patent for the purposes of this trial.

Hilton Davis also contends that Warner-Jenkinson has used manufacturing processes for making FD&C Red 40 and FD&C Yellow 6 food dyes which willfully infringe claims 1, 2, 3, 13, and 14 of the Hilton Davis patent.

Finally, Hilton Davis contends that Warner-Jenkinson is liable to it for its damages resulting from the unauthorized use of its patented food dye process.

Defendant Warner-Jenkinson contends that the Hilton Davis patent is invalid, and additionally, Warner-Jenkinson denies that the Warner-Jenkinson process for producing Red 40 and Yellow 6 infringes claims 1, 2, 3, 13 and 14 of the Hilton Davis patent because it uses different process conditions for ultrafiltering its dyes than those that are claimed by the plaintiff in the subject patent.

The claims of the patent define the limits of the invention. The claims of the patent must define the particular thing claimed to have been invented so that the public will know what the thing is and will, therefore, be able to avoid infringing upon the claims of the patent.

The function of a claim is to define the exact boundaries or limits of the claimed invention.

[2046] Neither the specification nor the drawings of a patent can be infringed. Only the patent claims can be invalid or infringed.

A patent may contain some claims which are valid and some claims which are invalid. A patent may also have some claims which are infringed and some claims which are not infringed. The only claims that you need to consider in this case are claims 1, 2, 3, 13 and 14 of the Hilton Davis patent.

Each of the claims of the patent is a separate definition of the invention. You should consider each claim on its own merits when evaluating both the issues of validity and infringement.

Each claim must be interpreted the same way both when considering its validity and when considering whether it is infringed.

In the Hilton Davis patent, claim 1 is known as an independent claim. All of the elements or steps required for the the process defined by the claim are written out in the claim itself.

The remaining claims in the Hilton Davis patent are known as dependent claims. Each of such dependent claims incorporates all of the elements or steps of claim 1 upon which it is dependent.

This means that the claims which are dependent on claim 1 are to be read just as if you were reading claim 1, [2047] then add into claim 1 the elements or steps recited in the dependent claims. Because if the dependent claims include all of the limitations in claim 1 from which they directly or indirectly depend, if you find that claim 1 from which the other claims depend is not infringed, then you must find that the remaining dependent claims, which are numbers 2, 3, 13 and 14, are likewise not infringed. You must determine if each of the independent and dependent claims is infringed.

The language of the Hilton Davis patent claims must be construed as it would be by those of ordinary skill in the art. In understanding the meaning of words used in the claims, you may consider the patent specification, the Prosecution history of the patent, other claims of the patent, testimony of expert witnesses, the circumstances surrounding the inception of the patent application and the meaning of words as contained in technical literature which are part of the legal evidence and the state—which are part of the legal evidence and the state of the prior art.

A patent granted by the Patent and Trademark Office is invalid if the claimed subject matter as a whole would have been obvious to a person of ordinary skill in the pertinent art at the time the patented invention was made. Patentability shall not be negated by the manner in which the invention was made.

In determining obviousness of the claimed subject [2048] matter of each of the claims of the patent in suit, the following steps should be taken by you: One, you should determine the scope and content of the prior art relied upon against the patent in suit. Two, you should then identify the difference, if any, between each claim of the patent in suit and the prior art. And three, you should determine the level of ordinary skill in the pertinent art at the time the invention of the patent in suit was made.

You must make each of the determinations as to of the time the Hilton Davis invention was made.

You should consider such secondary considerations as commercial success, long-felt but unresolved need, failure of others to solve the problem and acquiescence in the patent by others.

The defendant Warner-Jenkinson must prove invalidity because of obviousness by clear and convincing legal evidence.

During the course of this trial you have heard the term "prior art" used frequently. One of Warner-Jenkinson's assertions in this case is that there were certain prior public disclosures which constitute prior art within the meaning of the law. The term "prior art" includes that which was: One, known or used by others in this country before the date of invention by inventor; or

Two, patented or described in a printed publication in this or a foreign country before the date of invention; or

[2049] Three, patented or described in a printed publication in this or a foreign country for more than one year prior to the date of the application herein; or

Four, publicly used or on sale in this country more than one year prior to the date of the application therefor; or

Five, made or built by another person before the date of the invention where the thing made or built was not abandoned, suppressed or concealed.

The scope of prior art is defined as that reasonably pertinent to the particular problem with which the inventor was involved.

I previously instructed you that when the United States Patent Office grants a patent it is presumed that the patent examiner was correct. If a patent challenger contends that a patent is invalid but relies only upon the same prior art that the examiner considered in granting the patent, he asserts that the examiner was wrong, and the presumption requires that he prove this by clear and convincing legal evidence.

If the patent challenger relies only on prior art, which is not more pertinent or relevant than the art considered by the examiner, then such challenger must also prove invalidity by clear and convincing legal evidence.

If the patent challenger relies upon prior art which was not considered by the examiner, and which is more pertinent [2050] than that which was considered, then invalidity must still be proved by clear and convincing legal evidence, however, the patent challenger may have an easier time doing so.

In determining whether or not the claims of the patent in suit would have been obvious at the time, it is not necessary that there be absolute predictability of the result. It is only required that there be a reasonable probability the beneficial result will be achieved to show obviousness.

In reaching your determination on the issue of obviousness, you should consider whether the subject matter of the invention was also developed independently by other persons either before the alleged inventors of the patent in suit or about the same time.

It is fundamental in patent law that one who applies a known process to a known chemical does not thereby invent an unobvious patentable process unless it produces a different or an unexpected result over the prior art. If you find that defendant Warner-Jenkinson has proved by clear and convincing legal evidence that the process contained in the patent was an existing process, that plaintiff simply applied it to an existing chemical for ultrafiltration, and that the process failed to produce a different or unexpected result over the prior art, then you must find that the patented process is obvious and that the plaintiff's patent claims are invalid.

There have been frequent references to a person [2051] having ordinary skill in the art. Such a person is only hypothetical. It is that person who is presumed to be aware of all the pertinent prior art. The actual inventor's skill is irrelevant to the inquiry. A person of ordinary skill in the art is also presumed to be one who thinks along the line of conventional wisdom in the art.

To reach a proper conclusion, you must step backward in time and into the shoes worn by that hypothetical person when the invention was unknown and just before it was made. You must then determine, in light of all the legal evidence, whether the patent challenger has convincingly established that the claimed invention as a whole would have been obvious at the time to that hypothetical person.

You are also to determine the level of ordinary skill in the art to which the claimed invention pertains at the time the claimed invention was made. Factors to be considered in determining the level of ordinary skill in the pertinent art are: The educational level and years of ex-

perience of the person or persons who you find to have developed the processes that are the subject of this case and of others working in the pertinent art, the types of problems encountered in the art, the teachings of the prior art, patents, and publications of other persons or companies, and the sophistication of the technology.

One of the considerations of obviousness is the [2052] difference between the pertinent prior art and the claims of the Hilton Davis patent. You must consider the claimed invention as a whole.

The difference may seem slight, but it may also have been the key to success and advancement in the art resulting from the invention.

The specification of the patent must set forth the best mode contemplated by the inventor of carrying out his invention. The purpose of the best mode requirement is to restrain inventors from applying for a patent while at the same time concealing from the public preferred embodiments of their inventions which they have in fact conceived. Thus, the best mode inquiry focuses on the inventor's state of mind as of the time he filed his application. To prove that the best mode requirement was not satisfied, defendant must prove by clear and convincing legal evidence that the inventors knew of a better mode of carrying out the claimed invention than they disclosed in the specification of the patent, and, two, the inventors concealed, whether accidentally or intentionally, that better mode. What is required is an adequate disclosure of the best mode, not a guarantee that every aspect of the specification be precisely and universally reproducible. An inventor is not required to supply production specifications. Compliance with the best mode requirement exists when an inventor discloses his preferred embodiment. The fact that [2053] Hilton Davis may eventually have used a commercial process better than that disclosed in the patent is not pertinent to whether the specification disclosed the best

mode contemplated by the inventor in carrying out his invention.

You must look at all the circumstances at the time the application was filed. Finally, each claim must be considered individually for compliance with the best mode requirement. A finding that the best mode has not been satisfied only affects those claims covering subject matter which has not been disclosed in compliance with the best mode requirement.

If patent statute provides that whoever, without authority from the patent owner, makes, uses or sells any patented invention within the United States during the term of the patent, infringes the patent.

Infringement is determined by comparing the accused process with the invention set forth in the claims of the patent. Two inquiries are involved: A determination of the scope of the claims, and, two, whether the claimed invention has been infringed.

Hilton Davis asserts that the Warner-Jenkinson process for making food dyes infringes the Hilton Davis patent under the doctrine of equivalents. The doctrine of equivalents exists to prevent a fraud on the patent.

The concept of the doctrine of equivalents is [2054] designed to protect the patent holder from an unscrupulous infringer who appropriates the invention but avoids the literal language of the claims. In this regard, consideration must be given for the purpose for which a step is used in the claims of the patent and in defendant's processes and the functions which they perform.

You may find infringement under the doctrine of equivalents when the accused process and the claimed invention perform substantially the same function in substantially the same way to yield substantially the same result even though the processes differ in name, form or shape.

Hilton Davis must prove infringement under the doctrine of equivalents by a preponderance of the legal evidence.

Though application of the doctrine of equivalents extends the protection of the patent beyond the literal words contained in the claims, it is not proper to erase the meaningful limitations of the claims on which the public is entitled to rely in avoiding infringement, and you must look to the claims section of the patent to determine the coverage and limitations of the patent.

The doctrine of prosecution history estoppel precludes a patent owner from obtaining a claim construction through the application of the doctrine of equivalents that would resurrect subject matter surrendered during prosecution [2055] of a patent application. That is not to say, however, that whenever a limiting amendment or argument is made during prosecution, the patent owner loses all coverage between what the claims literally cover and what they would have covered prior to the amendment or argument. A close examination must be made as to, not only what was surrendered, but also the reason for such a surrender. The fact that claims were narrowed does not always mean that a patent owner is completely prohibited from recapturing some of what was originally claimed. Depending on the nature and purpose of an amendment, it may have a limiting effect within a spectrum ranging from great to small to zero. Determination of the effect on the doctrine of equivalents from actions taken before the PTO requires consideration of the nature of such actions, the reasons therefore, the prior art distinguished, and the examiner's objections thereby overcome.

A patent owner cannot obtain, under the doctrine of equivalents, coverage which could not have been obtained from the PTO by literal claims. In making this determination, you should consider whether the Hilton Davis patent claims as a whole, when interpreted to cover the Warner-Jenkinson's processes, "read on" the prior art.

If you find that Warner-Jenkinson's process infringes the Hilton Davis patent's claims, you must also decide whether or not plaintiff has proven by clear and [2056] convincing evidence that defendant's infringement was willful.

When a potential infringer has actual notice of another's patent rights, he has the duty to exercise due care to determine whether or not he is infringing.

There can be no willful infringement unless you find that plaintiff has proven that defendant knew or as a reasonable corporation should have known of plaintiff's patent and that defendant knew or as a reasonable corporation should have known that use of its process infringed plaintiff's patent claims.

Your decision concerning whether or not there is willful infringement must be based on the totality of the circumstances presented in this case. You may not find that any infringement was willful unless you find that plaintiff has proven that the defendant did not exercise due care to determine whether or not it was infringing the patent.

The duty of due care normally requires that a potential infringer obtain competent legal advice before infringing or continuing to infringe. The infringer must not only show an opinion from competent counsel but also that it had exercised reasonable and good faith adherence to the analysis and advice therein. The opinions of counsel must also be authoritative.

You should also consider whether defendant reasonably relied upon its counsel's opinion to determine [2057] whether defendant had a reasonable basis for believing that it had a right to use its process involved in this litigation.

You may consider that there was no willful infringement if you find that defendant has mounted a good faith and substantial challenge in this trial to the validity of the patent and to all claims of infringement. Since an invalid patent cannot be infringed, you should consider in your

determination of willfulness, the reasonableness of the defenses raised by defendant in connection with any of the defendant's assertions that plaintiff's patent is invalid independent of defendant's assertion relating to its non-infringement of plaintiff's patent.

In assessing whether plaintiff has proven that the defendant willfully infringed the Hilton Davis patent, you must look to all the circumstances in determining whether the defendant conducted itself as a reasonable corporation would conduct itself under the same or similar circumstances.

If you find that the Hilton Davis patent remains valid and has been infringed by Warner-Jenkinson, you must then determine an amount of damages, if any, to which Hilton Davis has proven to you by a preponderance of the legal evidence it is entitled.

In general, a patent owner is entitled to damages that are adequate to compensation for the infringement, but in no event less than a reasonable royalty. Such damages [2058] represent compensation for the pecuniary loss suffered from the infringement without regard to the question whether the defendant has gained or lost by his unlawful acts.

There are two methods by which damages may be calculated: the profits the patent owner lost from the infringement, or, if lost profits cannot be ascertained, then a reasonable royalty must be determined.

The amount of the damages need not been proven with unerring precision. It is enough if the legal evidence shows the extent of the damages to a reasonable certainty as a matter of just and reasonable inference, although the result be only approximate. When the amount of the damages cannot be ascertained with precision, any doubts regarding the amount must be resolved against the infringer.

A patent owner may recover lost profits by proving that, but for the infringement, the patent owner would have made the sales the infringer made, charged higher prices, or incurred lower expenses. The patent owner need not prove causation as an absolute certainty. Legal evidence showing a reasonable probability that the patent owner would have made the infringing sales the infringer made will suffice. The patent owner is not obligated to negate every possibility that a purchaser might not have bought the patent owner's product instead of the infringer's one, or might have foregone the purchase altogether.

[2059] To obtain as damages the profits on sales he would have made absent the infringement, that is, the sales made by the infringer, a patent owner must prove or may prove, one, demand for the patented product; two, absence of acceptable non infringing substitutes; three, his manufacturing and marketing capability to exploit the demand; and four, the amount of the profits he would have made. These elements, however, are not an absolute requirement in every case.

The factors to be considered in calculating lost profits, if any, include the amount or number of lost sales, the gross receipts the patent owner would obtain would have obtained from the lost sales had there been no infringement, the cost of sales to be deducted from gross receipts, and the patent owner's profit on the lost sales.

To be deemed acceptable, the alleged acceptable non-infringing substitute must not have a disparately higher price than or process characteristics significantly different from the product produced by the patented process.

As noted earlier, a reasonable royalty is to be awarded as damages only if the patent owner cannot establish its lost profits. The purposes of reasonable royalty is to set a floor below which the damages are not authorized to go. The objective of a reasonable royalty is to place the patent

owner in at least as good a position as he would have been had the alleged infringer entered into a reasonable royalty agreement. [2060] A reasonable royalty is to be determined as of the date when the infringement, if any, began.

A reasonable royalty is an amount which a person, desiring to manufacture, use or sell a patented item or article, as a business proposition, would be willing to pay as a royalty and yet still be able to make and sell the patented article in the market at a reasonable profit. It is a hypothetical royalty resulting from arms length negotiations between two companies, although there is no actual willingness on either side to enter into a royalty agreement. An alleged infringer's failure to have made a profit from its alleged infringement and its willingness to pay a royalty are irrelevant.

You will have with you in the jury room the following special verdicts. No inference is to be drawn from the way I read the verdict forms and you must unanimously agree upon your verdict. And since we have already gone through them, I will not read them again to you. I have placed the verdict form in the front part of the notebook.

Nothing that I have said in these instructions and nothing in the manner in which the special verdicts have been prepared or explained to you is intended to suggest or convey in any way a result I think you should reach, as this is the exclusive duty and responsibility of the jury. I state to you categorically that the Court has no opinion as to the disputed [2061] facts of this case or the propriety of any verdict you must return.

I cannot embody all the law in any single part of these instructions. In considering one portion, you must consider it in the light of and in harmony with all the instructions.

I have instructed you on all the law necessary for your deliberations. Whether certain instructions are applicable may depend upon the conclusions you reach on the facts.

It is your duty, as jurors, to confer with one another, and to deliberate with a view to reaching an agreement, if you can do so without doing violence to your individual judgment. Each of you must decide the case for yourself, but do so only after an impartial consideration of all the legal evidence in the case with other jurors. In the course of your deliberations, do not hesitate to re-examine your own views and change your opinion if you are convinced it is erroneous.

Do not surrender your honest conclusion as to the weight or effect of the legal evidence, however, solely because of the opinion of other jurors or for the mere purpose of returning a verdict.

Remember always that you are not partisans. You are judges, impartial triers of the facts. Your sole interest is [2062] to ascertain the truth from the legal evidence in the case. Do not take a firm position at the outset and then be too proud to change your position if you become convinced that your position is wrong.

You must not be influenced by any consideration of sympathy or prejudice. It is your duty to carefully weigh the legal evidence, to decide all disputed questions of fact, to apply these instructions to your findings, and to render your verdict accordingly. In fulfilling your duty, your efforts must be to arrive at a just verdict. Consider the legal evidence and make your findings with intelligence and impartiality, and without bias, sympathy or prejudice, so that the litigants will feel that their case was fairly and impartially tried. If, during the course of the trial, I said or did anything which you consider an indication of my view on any disputed fact, you are instructed to disregard it, because only you, the jury, determine such matters.

If during your deliberations you wish to communicate with me, please put your communication in writing and give it to the Courtroom Clerk who will deliver it to me.

The Court will place into your possession the exhibits, the special verdicts and a copy of these instructions.

Upon retiring to the jury room, you will select one of your number to act as your foreperson. The foreperson will [2063] retain possession of these records and return them to the courtroom. The foreperson will see that your discussions are orderly and that each juror has the opportunity to discuss the case and to cast his or her vote. Otherwise, the authority of the foreperson is the same as any other juror.

Until your special verdict is announced in open court, you are not to disclose to anyone else the status of your deliberations or the nature of your verdict.

Now, ladies and gentlemen of the jury, is there anyone present on the panel that feels that they could not enter into deliberations in this matter and so far as you know not reach a fair and impartial verdict in this case, judgment in this case? I take it by your silence that there is no reason. If you'll excuse me just a minute.

* * * *

[Filed Jun. 16, 1992]

UNITED STATES DISTRICT COURT
SOUTHERN DISTRICT OF OHIO
WESTERN DIVISION

C-1-91-218

HILTON DAVIS CHEMICAL CO.,
Plaintiff

v.

WARNER-JENKINSON COMPANY, INC.,
Defendant

SPECIAL VERDICTS

1. We, the Jury, unanimously find that defendant Warner-Jenkinson has proved by clear and convincing legal evidence that the following claims of the Hilton Davis patent are invalid because the differences between such claim and the pertinent prior art were such that the subject matter of the claim as a whole would have been obvious to a person having ordinary skill in the pertinent art at the time of the invention:

Claim 1	No (valid) <u>X</u>	Yes (invalid) _____
Claim 2	No (valid) <u>X</u>	Yes (invalid) _____
Claim 3	No (valid) <u>X</u>	Yes (invalid) _____
Claim 13	No (valid) <u>X</u>	Yes (invalid) _____
Claim 14	No (valid) <u>X</u>	Yes (invalid) _____

2. We, the Jury, unanimously find that defendant Warner-Jenkinson has proved by clear and convincing legal evidence that the following claims of the Hilton Davis patent are invalid because employees of Osmonics, rather than Drs. Robert W. J. Rebhahn and Wayne L. Cook, invented a portion or all of the claim:

Claim 1 No (valid) X Yes (invalid) _____
 Claim 2 No (valid) X Yes (invalid) _____
 Claim 3 No (valid) X Yes (invalid) _____
 Claim 13 No (valid) X Yes (invalid) _____
 Claim 14 No (valid) X Yes (invalid) _____

3. We, the Jury, unanimously find that defendant Warner-Jenkinson has proved by clear and convincing legal evidence that the following claims of the Hilton Davis patent are invalid because they apply an existing process to existing chemicals which does not produce a different or an unexpected result over the prior art:

Claim 1 No (valid) X Yes (invalid) _____
 Claim 2 No (valid) X Yes (invalid) _____
 Claim 3 No (valid) X Yes (invalid) _____
 Claim 13 No (valid) X Yes (invalid) _____
 Claim 14 No (valid) X Yes (invalid) _____

4. We, the Jury, unanimously find that defendant Warner-Jenkinson has proved by clear and convincing legal evidence that the following claims of the Hilton Davis patent are invalid because another person was not named as a coinventor to such claim:

Claim 1 No (valid) X Yes (invalid) _____
 Claim 2 No (valid) X Yes (invalid) _____
 Claim 3 No (valid) X Yes (invalid) _____
 Claim 13 No (valid) X Yes (invalid) _____
 Claim 14 No (valid) X Yes (invalid) _____

5. We, the Jury, unanimously find that defendant Warner-Jenkinson has proved by clear and convincing legal evidence that the Hilton Davis patent is invalid because the inventors knew of and concealed a better mode of carrying

out the invention than was set forth in the specification of the patent.

NO (valid) X YES (invalid) _____

6. We, the Jury, unanimously find that plaintiff Hilton Davis has proved by a preponderance of the legal evidence that defendant Warner-Jenkinson has infringed upon the following claims of the Hilton Davis patent:

Claim 1 Yes (infringed) X No (not infringed) _____
 Claim 2 Yes (infringed) X No (not infringed) _____
 Claim 3 Yes (infringed) X No (not infringed) _____
 Claim 13 Yes (infringed) X No (not infringed) _____
 Claim 14 Yes (infringed) X No (not infringed) _____

7. As to any claim found to be infringed in Special Verdict No. 6, We, the Jury, unanimously find that plaintiff Hilton Davis has proved by clear and convincing legal evidence that defendant Warner-Jenkinson acted willfully to infringe that claim of the patent:

Claim 1 Yes (willful) _____ No (not willful) X
 Claim 2 Yes (willful) _____ No (not willful) X
 Claim 3 Yes (willful) _____ No (not willful) X
 Claim 13 Yes (willful) _____ No (not willful) X
 Claim 14 Yes (willful) _____ No (not willful) X

8. We, the Jury, unanimously find by a preponderance of the legal evidence that Hilton Davis has proved that the reasonable royalty for the use of the Hilton Davis patent by Warner-Jenkinson is:

\$887,767.00

9. We, the Jury, unanimously find that plaintiff Hilton Davis has proved by a preponderance of the legal evidence that it lost profits as a result of the infringement of its patent by defendant Warner-Jenkinson in the amount of:

\$3,564,705.00

and award to plaintiff Hilton Davis against the defendant Warner-Jenkinson damages in the sum of:

\$3,564,705.00

[juror signature]

[juror signature]

[juror signature]

[juror signature]

[juror signature]

[juror signature]

[juror signature]

[juror signature]

Date 6/16/92

[PATENT FILE]

REGULAR UTILITY

481038

Serial Number 06/481,038

Filing Date 03/28/83

Class 260

Subclass 208

Group Art Unit 124

Examiner Higel

Applicants

Robert W. J. Rebhahn, West Chester, OH; Wayne L. Cook, Cincinnati, OH.

Continuing Data Verified

Foreign/Pct Applications Verified

Foreign Filing License Granted 04/25/83

* * * *

As Filed

State or Country OH

Sheets Drwgs. 0

Total Claims 13

Indep. Claims 1

Filing Fee Received \$300.00

Attorney's Docket No. 7364

Address

H. Woodrow Wyatt
Sterling-Winthrop Research Institute
Rensselaer, NY 12144

Title

Process for Purification of Azo Dyes Useful in Food-stuffs

This is to certify that annexed hereto is a true copy from the records of the United States Patent and Trademark Office of the File Wrapper and Contents of the file identified above.

By authority of the
COMMISSIONER OF PATENTS AND TRADEMARKS

/s/

Certifying Officer

Dated Apr. 3, 1992

[Application]

REGULAR UTILITY

ABANDON

481038

Serial Number 06/481,038

Filing Date 03/28/83

Class 260

Subclass 208

Group Art Unit 124

Examiner Higel

Applicants

Robert W. J. Rebhahn, West Chester, OH; Wayne L.
Cook, Cincinnati, OH.

Continuing Data Verified

Foreign/Pct Applications Verified

Foreign Filing License Granted 04/25/83

* * * *

As Filed

State or Country OH

Sheets Drwgs. 0

Total Claims 13

Indep. Claims 1

Filing Fee Received \$300.00

Attorney's Docket No. 7364

Address

H. Woodrow Wyatt
Sterling-Winthrop Research Institute
Rensselaer, NY 12144

Title

Process for Purification of Azo Dyes Useful in Food-
stuffs

* * * *

PROCESS FOR PURIFICATION OF AZO DYES USEFUL IN FOODSTUFFS

BACKGROUND OF THE INVENTION

(a) Field of the Invention

This invention relates to the field of purification, by ultrafiltration techniques, of dyes of the azo class useful in foodstuffs.

(b) Information Disclosure Statement

Bollenback et al. U.S. Patent 3,249,444, patented May 3, 1966, describes an ultrafiltration process for increasing the tinctorial power of caramel color in which sugar, i.e. uncaramelized sugar, is separated from caramel color by ultrafiltration through a semi-permeable membrane which permits passage of small, uncolored molecules in solutions containing caramel color and rejects the passage of larger, polymeric caramel color molecules, thus enhancing the color of the concentrate. Preferred membranes for the process are made of vinyl plastics, and preferred pressures are in the range from 20 to 100 p.s.i.g.

Adams et al. U.S. Patent 3,544,455, patented December 1, 1970, discloses a process for the purification of itaconic acid by reverse osmosis through a semi-permeable membrane composed of cellulose acetate or polyamide in which itaconic acid and water are forced to the downstream side of the membrane, while inorganic salts, colored materials and organic materials remain on the upstream side. The process is carried out under a hydrostatic pressure of from 100 to 1,000 p.s.i.g. and at a pH in the range from 2 to 4.

* * * *

The membranes used in the practice of the present process, manufactured by Osmonics, Inc. of Minnetonka,

Minnesota, and generally referred to as reverse osmosis/ultrafiltration membranes, are generally formulated of cellulose acetate, and in the process of the present invention, have a nominal pore diameter of 11 Angstroms. The filtration is carried out under a hydrostatic pressure of 200 to 400 p.s.i.g. applied to the upstream side of the membrane. By use of a membrane having the appropriate critical pore size, the impurities, along with a large quantity of water, are thus forced through the membrane and accumulate on the downstream side as the permeate, while the desired product molecules are rejected by the membrane and remain on the upstream side of the membrane where the product becomes more and more concentrated as more and more water and impurities are forced to the downstream side.

* * * *

In carrying out the present process, the reaction mixture as produced in the diazo coupling and as fed to the ultrafiltration unit, generally has a pH of around 9.0. While these solutions can be subjected successfully to ultrafiltration, it is preferred to adjust the pH to around 7.0 to 8.0 before passage through the ultrafiltration membrane.

* * * *

We claim:

1. In a process for the purification of an azo dye selected from the group consisting of the disodium salts of 1-[(6-methoxy-4-sulfo-3-methylphenyl)azo]-2-naphthol-6-sulfonic acid and 1-[(4-sulfophenyl)azo]-2-naphthol-6-sulfonic acid as the products resulting, respectively, from the diazotization of 5-methoxy-2-methyl sulfanilic acid or sulfanilic acid in water with sodium nitrite in the presence of hydrochloric acid followed by the coupling under alkaline conditions of the resulting respective, 5-methoxy-4-sulfo-2-methylphenyldiazonium chloride or 4-sulfophenyldiazonium chloride with sodium 2-naphthol-6-sulfonate, said azo dye being present in the resulting reaction mixtures, along with impurities, the improvement which com-

prises: directly subjecting the reaction mixture resulting from said coupling to ultrafiltration through a membrane composed of cellulose acetate having a nominal pore diameter of 11 Angstroms under a hydrostatic pressure of from about 200 to 400 p.s.i.g. to thereby cause separation of said impurities from said azo dye, said impurities passing into the permeate on the downstream side of said membrane and said azo dye remaining in the concentrate, and when substantially all said impurities have been removed from said concentrate, as evidenced by their essential absence in said permeate, recovering said azo dye, in greater than 90% purity and in a yield of around 98% of the total available color produced in said process, from said concentrate by evaporation of said concentrate to dryness.

2. A process according to Claim 1 wherein the concentration of the azo dye in said concentrate is maintained at around 25% (w/w) by recycling the concentrate and addition of water thereto.

3. A process according to Claim 1 wherein ultrafiltration is carried out until the concentration of the azo dye in the concentrate is maintained at around 5% (w/w).

4. A process according to Claim 2 wherein the pH of the reaction mixture is adjusted to 7.0 to 8.0 before ultrafiltration.

5. A process according to Claim 3 wherein the pH of the reaction mixture is adjusted to 7.0 to 8.0 before ultrafiltration.

* * * *

[Examiner's Rejection]

UNITED STATES DEPARTMENT OF COMMERCE

Patent and Trademark Office

Address: Commissioner of Patents and Trademarks
Washington, D.C. 20231

Serial Number 06-481-038

Filing Date 03/31/83

First Named Applicant Rebhahn

Attorney Docket No. 7364

H. Woodrow Wyatt
Sterling-Winthrop Research Institute
Rensselaer, NY 12144

Examiner Higel, F.

Art Unit 124

Paper Number

Date Mailed: 10-02-84

This is a communication from the examiner in charge of
your application.

Commissioner of Patents and Trademarks

☒ This application has been examined☐ Responsive to communication filed on _____☐ This action is made final.

A shortened statutory period for response to this action is set to expire three month(s) from the date of this letter. Failure to respond within the period for response will cause the application to become abandoned. 35 U.S.C. 133

Part I THE FOLLOWING ATTACHMENT(S) ARE
PART OF THIS ACTION:

1. ☒ Notice of References Cited by Examiner, PTO-892.
2. ☐ Notice re Patent Drawing, PTO-948
3. ☒ Notice of Art Cited by Applicant, PTO-1449
4. ☐ Notice of Informal Patent Application, Form PTO-152
5. ☐ Information on How to Effect Drawing Changes, PTO-1474
6. ☐ _____

Part II SUMMARY OF ACTION

1. ☒ Claims 1 to 13 are pending in the application.
Of the above, claims _____
_____ are withdrawn from consideration.
2. ☐ Claims _____ have been cancelled.
3. ☐ Claims _____ are allowed.
4. ☒ Claims 1 to 13 are rejected.
5. ☐ Claims _____ are objected to.
6. ☐ Claims _____ are subject to restriction or election requirement.
7. ☐ This application has been filed with informal drawings which are acceptable for examination purposes until such time as allowable subject matter is indicated.
8. ☐ Allowable subject matter having been indicated, formal drawings are required in response to this Office action.
9. ☐ The corrected or substitute drawings have been received on _____. These drawings are

- ☐ acceptable;
- ☐ not acceptable (see explanation).
10. ☐ The ☐ proposed drawing correction and/or the ☐ proposed additional or substitute sheet(s) of drawings, filed on _____, has (have) been
- ☐ approved by the examiner.
- ☐ disapproved by the examiner (see explanation).
11. ☐ The proposed drawing correction, filed _____, has been
- ☐ approved.
- ☐ disapproved (see explanation). However, the Patent and Trademark Office no longer makes drawing changes. It is now applicant's responsibility to ensure that the drawings are corrected. Corrections MUST be effected in accordance with the instructions set forth on the attached letter "INFORMATION ON HOW TO EFFECT DRAWING CHANGES", PTO-1474.
12. ☐ Acknowledgment is made of the claim for priority under 35 U.S.C. 119. The certified copy has
- ☐ been received
- ☐ not been received
- ☐ been filed in parent application, serial no. _____; filed on _____.
13. ☐ Since this application appears to be in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under Ex parte Quayle. 1935 C.D. 11; 453 O.G. 213.
14. ☐ Other

EXAMINER'S ACTION

Serial No. 481038

Art Unit 124

The title of the invention is not descriptive. A new title is required that is clearly indicative of the invention to which the claims are directed.

The lengthy specification has not been checked to the extent necessary to determine the presence of all possible minor errors. Applicant's cooperation is requested in correcting any errors of which applicant may become aware in the specification. Receipt is acknowledged of the information Disclosure Statement filed May 26, 1984, which has been entered in the file.

The following is a quotation of 35 U.S.C. 103 which forms the basis for all obviousness rejections set forth in this Office action:

A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negated by the manner in which the invention was made.

Claims 1 to 11 are rejected under 35 U.S.C. 103 as being unpatentable over Booth et al, cited. The reference discloses the purification of azo dyes by ultrafiltration in high purity and yield. It would be obvious to purify azo dyestuffs by the method of the reference in the absence of any unobvious results.

Claims 12 and 13 are rejected under 35 U.S.C. 103 as being unpatentable over Colour Index, in view of Booth et al. The purified claimed compound or obvious variations thereof would be obvious from the compounds of Colour Index purified by the method of Booth et al, in the absence of any obvious or unexpected properties.

Bollenback et al, Adams et al, Teed et al, EPO, South African, Osmonics and Spatz I and II, cited by applicants, show the state of the art.

No claim is allowed.

Any inquiry concerning this communication should be directed to Floyd D. Higel at telephone number 703-557-3920.

/s/ Floyd D. Higel
FLOYD D. HIGEL
Primary Patent Examiner
Group Art Unit 124

FDHigel:cvm

A/C 703

557-3920

9/13/84

U.S. Department of Commerce
Patent and Trademark Office

NOTICE OF REFERENCES CITED

Serial No. 481,038

Group Art Unit 124

Attachment To Paper Number 3

Applicant(s) Robert W.J. Rebhahn et al

U.S. PATENT DOCUMENTS

Document No. 4189380

Date 02-1980

Name Booth et al

Class 260

Subclass 144X

Filing Date If Appropriate

* * * *

FOREIGN PATENT DOCUMENTS

* * * *

OTHER REFERENCES

(Including Author, Title, Date, Pertinent Pages, Etc.)

Colour Index, 8rd Bd., Volume 4, page 4087

#15985 (1971)

* * * *

Examiner

/s/ Floyd D. Higel

Date 09/07/84

A copy of this reference is not being furnished with
this office action.

(See Manual of Patent Examining Procedure,
section 707.05(a).)

[Notice of Abandonment]

UNITED STATES DEPARTMENT OF COMMERCE

Patent and Trademark Office

Address: Commissioner of Patents and Trademarks
Washington, D.C. 20231

Serial Number 06/481,038

Filing Date 03/31/83

First Named Applicant Rebhahn

Attorney Docket No. 7364

H. Woodrow Wyatt
Sterling-Winthrop Research Institute
Rensselaer, NY 12144

Examiner Higel, F

Art Unit 124

Paper Number 4

Date Mailed: 05/06/85

NOTICE OF ABANDONMENT

This application is abandoned in view of:

1. ☒ Applicant's failure to respond to the Office letter, mailed October 02, 1984.
 2. ☐ Applicant's letter of express abandonment which is in compliance with 37 C.F.R. 1.138.
 3. ☐ Applicant's failure to timely file the response received _____ within the period set in the Office letter.
 4. ☐ Applicant's failure to pay the required issue fee within the statutory period of 3 months from the mailing date of _____ of the Notice of Allowance.
- ☐ The issue fee was received on _____.

- ☐ The issue fee has not been received in Allowed Files Branch as of _____.

In accordance with 35 U.S.C. 151, and under the provisions of 37 C.F.R. 1.316(b), applicant(s) may petition the Commissioner to accept the delayed payment of the issue fee if the delay in payment was unavoidable. The petition must be accompanied by the issue fee, unless it has been previously submitted, in the amount specified by 37 C.F.R. 1.17 (1), and a verified showing as to the causes of the delay.

If applicant(s) never received the Notice of Allowance, a petition for a new Notice of Allowance and withdrawal of the holding of abandonment may be appropriate in view of *Delgar Inc. v. Schuyler*, 172 U.S.P.Q. 513.

5. ☐ Applicant's failure to timely correct the drawings and/or submit new or substitute formal drawings by _____ as required in the last Office action.
- ☐ The corrected and/or substitute drawings were received on _____.
6. ☐ The reason(s) below.

/s/ Floyd D. Higel
FLOYD D. HIGEL
Primary Patent Examiner
Group Art Unit 124

[Continuation in Part Application]

Serial Number 677118
 Patent Date Dec. 24, 1985
 Patent Number 4560746
 Serial Number 06/677,118
 Filing Date 11/30/84
 Class 260
 Subclass 208
 Group Art Unit 136 [124?]
 Examiner Higel

APPLICANTS

Robert W. J. Rebhahn, Berkeley, MA;
 Wayne L. Cook, Cincinnati, OH.

Continuing Data Verified

/s/ _____

This Appln is a CIP of 06/481,038 03/28/83, AB

/s/ Higel AU124

Foreign/Pct Applications Verified

/s/ _____

Foreign Filing License Granted 12/26/84

Foreign priority claimed

☐ Yes ☒ No

35 USC 119 conditions met

☐ Yes ☒ No

Verified and Acknowledged /s/ _____ Examiners Initials

As filed

State or Country NA

Sheets Drwgs. 0

Total Claims 17

Indep. Claims 1
 Filing Fee Received \$300.00
 Attorney's Docket No. 73648

ADDRESS

B. Woodrow Wyatt
 Sterling-Winthrop Research Institute
 Rensselaer, NY 12144

TITLE

Ultrafiltration Process For Purification of Dyes useful In Foodstuffs

PARTS OF APPLICATION FILED SEPARATELY

PREPARED FOR ISSUE

/s/ D. Washington
 (Docket Clerk)

AT ALLOWANCE

Sheets Drwgs None
 Figures Drwgs None
 Claims 17
 Class 534
 Subclass 840

Examined and Passed for Issue
 Floyd D. Higel
 Primary Patent Examiner
 Group Art Unit 124

RETENTION LABEL 168

Estimates of printed pages Issue fee due (est.) \$500
 Drawing(s)
 Space(s)
 Notice of allowance and issue fee due (est.)
 Date mailed 6/26/85
 Date paid 9-27-85

PROCESS FOR PURIFICATION OF DYES USEFUL IN FOODSTUFFS

RELATED APPLICATION

This is a continuation-in-part of our prior, copending application Serial No. 481,038, filed March 28, 1983, *now abandoned*.

* * * *

The membranes used in the practice of the present process, and generally referred to as reverse osmosis/ultrafiltration membranes, have a nominal pore diameter of 5-15 Angstroms, a preferred range being from 7-11 Angstroms. Membranes useful in the practice of the present invention are manufactured by Osmonics Inc. of Minnetonka, Minnesota or by the Celanese Corporation and are generally formulated of cellulose acetate, polyamide or polyvinylfluoride. The filtration is carried out under a hydrostatic pressure of approximately 200 to 400 p.s.i.g. applied to the upstream side of the membrane. By use of a membrane having the appropriate critical pore size, those impurities of a molecular size smaller than the nominal pore diameter of the membrane, along with a large quantity of water, are thus forced through the membrane and accumulate on the downstream side as the permeate, while the desired product molecules, as well as impurities of a molecular size larger than the nominal pore diameter of the membrane, are rejected by the membrane and remain on the upstream side thereof where the product becomes more and more concentrated as more and more water and impurities are forced to the downstream side.

* * * *

In carrying out the present process the reaction mixture, as produced in the diazo coupling and as fed to the ultrafiltration unit, generally has a pH of approximately 9.0. While these solutions can be subjected successfully

to ultrafiltration, it is preferred to adjust the pH to approximately 6.0 to 8.0 before passage through the ultrafiltration membrane.

* * * *

We claim:

1. In a process for the purification of a dye selected from the group consisting of the disodium salt of 1-[(6-methoxy-4-sulfo-3-methylphenyl)azo]-2-naphthol-6-sulfonic acid, the disodium salt of 1-[(4-sulfophenyl)azo]-2-naphthol-6-sulfonic acid, the trisodium salt of 1-[1-(4-sulfonaphthyl)azo]-2-naphthol-3,6-disulfonic acid, the disodium salt of 2-[1-(4-sulfonaphthyl)azo]-1-naphthol-4-sulfonic acid and the sodium salt of 2-(2-quinolyl)-1,3-indanedione-sulfonic acid as the products resulting, respectively, from the diazotization of 5-methoxy-2-methylsulfanilic acid in water with sodium nitrite in the presence of hydrochloric acid followed by the coupling under alkaline conditions of the resulting 5-methoxy-4-sulfo-2-methylphenyldiazonium chloride with sodium 2-naphthol-6-sulfonate; the diazotization of sulfanilic acid in water with sodium nitrite in the presence of hydrochloric acid followed by the coupling under alkaline conditions of the resulting 4-sulfophenyldiazonium chloride with sodium 2-naphthol-6-sulfonate; the diazotization of 4-aminonaphthalene-1-sulfonic acid in water with sodium nitrite in the presence of hydrochloric acid followed by the coupling under alkaline conditions of the resulting 1-sulfonaphthyl-4-diazonium chloride with disodium 2-naphthol-3,6-disulfonate; the diazotization of 4-aminonaphthalene-1-sulfonic acid in water with sodium nitrite in the presence of hydrochloric acid followed by the coupling under alkaline conditions of the resulting 1-sulfonaphthyl-4-diazonium chloride with sodium 1-naphthol-4-sulfonate; and the condensation of 2-quinaldine with phthalic anhydride followed by sulfonation of the resulting 2-(2-quinolyl)-1,3-indanedione, said dye being present in the resulting reaction mixtures, along with impurities, the improvement which comprises: subjecting an aqueous solution of the reaction

mixture resulting from said coupling or said sulfonation to ultrafiltration through a membrane having a nominal pore diameter of 5-15 Angstroms under a hydrostatic pressure of approximately 200 to 400 p.s.i.g. *at a pH from approximately 6.0 to 9.0*, to thereby cause separation of said impurities from said dye, said impurities of a molecular size smaller than the nominal pore diameter passing into the permeate on the downstream side of said membrane and said dye remaining in the concentrate, and when substantially all said impurities have been removed from said concentrate, as evidenced by their essential absence in said permeate, recovering said dye, in approximately 90% purity from said concentrate by evaporation of said concentrate to dryness.

2. A process according to Claim 1 wherein said membrane is composed of cellulose acetate, polyamide or polyvinylfluoride.

3. A process according to Claim 2 wherein the concentration of the dye in said concentrate is maintained at approximately 5-25% (w/w) by recycling the concentrate and adding water thereto.

4. A process according to Claim 2 wherein ultrafiltration is carried out until the concentration of the azo dye in the concentrate is maintained at approximately 5% (w/w).

5. A process according to Claim 3 wherein the pH of the reaction mixture is adjusted to 6.0 to 8.0 before ultrafiltration.

6. A process according to Claim 4 wherein the pH of the reaction mixture is adjusted to 6.0 to 8.0 before ultrafiltration.

* * * *

[Examiner's Rejection]

UNITED STATES DEPARTMENT OF COMMERCE
Patent and Trademark Office

Address Commissioner of Patents and Trademarks
Washington, DC 20231

Serial Number 06/677-118

Filing Date 11/30/84

First Named Applicant REBHahn

Attorney Docket No.

B. Woodrow Wyatt
Sterling-Winthrop Research Institute
Rensselaer, NY 12144

Examiner HIGEL, F

Art Unit 124

Paper Number 3

Date Mailed: 02/14/85

This is a communication from the examiner in charge
of your application.

COMMISSIONER OF PATENTS AND TRADEMARKS

☒ This application has been examined ☐ Responsive to communication filed on _____ ☐ This action is not. A shortened statutory period for response to this action is set to expire THREE month(s) from the date of this letter. Failure to respond within the period for response will cause the application to become abandoned. 35 U.S.C. 133

Part I THE FOLLOWING ATTACHMENT(S) ARE
PART OF THIS ACTION:

1. ☒ Notice of Reference Cited by Examiner, PTO-892.

2. ☐ Notice re Patent Drawing, PTO-948.
3. ☐ Notice of Art Cited by Applicant, PTO-1449
4. ☐ Notice of Informal Patent Application, Form
5. ☐ Information on How to Effect Drawing Changes, PTO-1474
6. ☐ _____

Part II SUMMARY OF ACTION

1. ☒ Claims 1 to 17 are pending in the application.

Of the above, claims _____ are withdrawn from consideration.

2. ☐ Claims _____ have been cancelled.
3. ☐ Claims _____ are allowed.
4. ☒ Claims 1 to 17 are rejected.
5. ☐ Claims _____ are objected to.
6. ☐ Claims _____ are subject to restriction or election requirement.
7. ☐ This application has been filed with informal drawings which are acceptable for examination purposes until such time as allowable subject matter is indicated.
8. ☐ Allowable subject matter having been indicated, formal drawings are required in response to this Office action.
9. ☐ The corrected or substitute drawings have been received on _____. These drawings are ☐ acceptable; ☐ not acceptable (see explanation).
10. ☐ The ☐ proposed drawing correction and/or the ☐ proposed additional or substitute sheet(s) of drawings, filed on _____ has (have) been ☐

approved by the examiner. ☐ disapproved by the examiner (see explanation).

11. ☐ The proposed drawing correction, filed _____, has been ☐ approved. ☐ disapproved (see explanation). However, the Patent and Trademark Office no longer makes drawing changes. It is now applicant's responsibility to ensure that the drawings are corrected. Corrections MUST be effected in accordance with the instructions set forth on the attached letter "INFORMATION ON HOW EFFECT DRAWING CHANGES", PTO-1474.
12. ☐ Acknowledgment is made of the claim for priority under 35 U.S.C. 119. The certified copy has ☐ been received ☐ not been received ☐ been filed in parent application, serial no. _____, filed on _____.
13. ☐ Since this application appears to be in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under Ex parte Quayle, 1935 C.D. 11; 453 O.G. 213.
14. ☐ Other

EXAMINER'S ACTION

Serial No. 677118

Art Unit 124

The title of the invention is not descriptive. A new title is required that is clearly indicative of the invention to which the claims are directed.

The lengthy specification has not been checked to the extent necessary to determine the presence of all possible minor errors. Applicant's cooperation is requested in correcting any errors of which applicant may become aware in the specification.

Receipt is acknowledged of the information Disclosure Statement filed November 30, 1984, which has been entered in the file.

The following is a quotation of 35 USC 103 forth in this Office action:

A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negated by the manner in which the invention was made.

Claims 1 to 17 are rejected under 35 USC 103 as being unpatentable over Booth et al, cited. The reference discloses the purification of azo dyes by ultrafiltration in high purity and yield. It would be obvious to purify azo dyestuffs by the method of the reference in the absence of any obvious results.

Claims 1 to 17 are rejected under 35 U.S.C. 103 as being unpatentable over Colour Index in view of Booth et al. It would be obvious to purify the compounds of

Colour Index by the method of Booth et al, in the absence of any obvious or unexpected results.

Bollenback et al, Adams et al, Teed et al, EPO, South Africa, Osmonics and Spatz I and II, cited by applicants, show the state of the art.

No claim is allowed.

Any inquiry concerning this communication should be directed to Floyd D. Higel at telephone number 703-557-3920.

/s/ Floyd D. Higel
FLOYD D. HIGEL
Primary Patent Examiner
Group Art Unit 124

Higel:tgh

AC 703

557-3920

2-8-85

[Examiner Interview Summary Record]

UNITED STATES DEPARTMENT OF COMMERCE
Patent and Trade Office

Address: Commissioner of Patents and Trademarks
Washington, D.C. 20231

Serial Number 677,118

Filing Date 11/30/84

First Named Applicant Robert W.J. Rebhahn

Attorney Docket No. 7364B

Examiner F D Higel

Art Unit 124

Paper Number 4

Date Mailed:

EXAMINER INTERVIEW SUMMARY RECORD

All participants (applicant, applicant's representative,
PTO personnel):

(1) William G. Webb

(2) Floyd D. Higel

(3) _____

(4) _____

Date of interview _____

Type: ☐ Telephonic ☒ Personal (copy is given to
☐ applicant ☒ applicant's representative).

Exhibit shown or demonstration conducted:

☐ Yes ☒ No. If yes, brief description: _____

Agreement

☐ was reached with respect to some or all of the
claims in question.

☒ was not reached.

Claims discussed: 1 to 17

Identification of prior art discussed:

Booth and Colour Index.

Description of the general nature of what was agreed to if an agreement was reached, or any other comments: Applicants' representative pointed out the 4 major differences between the claimed process and that of the Booth Patent. The examiner stated that if claim 1 were amended to contain the pH range of 6 to 9, the rejection on prior art would be overcome.

(A fuller description, if necessary, and a copy of the amendments, if available, which the examiner agreed would render the claims allowable must be attached. Also, where no copy of the amendments which would render the claims allowable is available, a summary thereof must be attached.)

Unless the paragraphs below have been checked to indicate to the contrary, A FORMAL WRITTEN RESPONSE TO THE LAST OFFICE ACTION IS NOT WAIVED AND MUST INCLUDE THE SUBSTANCE OF THE INTERVIEW (e.g., items 1-7 on the reverse side of this form). If a response to the last Office action has already been filed, then applicant is given one month from this interview date to provide a statement of the substance of the interview.

- ☐ It is not necessary for applicant to provide a separate record of the substance of the interview.
- ☐ Since the examiner's interview summary above (including any attachments) reflects a complete response to each of the objections, rejections and requirements that may be present in the last Office action, and since the claims are now allowable, this

completed form is considered to fulfill the response requirements of the last Office action.

/s/ Floyd D. Higel
Examiner's Signature

[Responsive Amendment]

IN THE UNITED STATES PATENT AND
TRADEMARK OFFICE

Group Art Unit 124

Examiner: F. D. Higel

IN RE PATENT APPLICATION OF
ROBERT W. J. REBHahn *et al.*

Serial No. 677,118

Filed November 30, 1984

For: Process for Purification of Dyes Useful in Foodstuffs

RESPONSIVE AMENDMENT

Hon. Commissioner of Patents and Trademarks
Washington, D.C. 20231

Sir:

In response to the Official Action mailed February 14, 1985, please amend the above-identified application as follows:

Change the title to read—ULTRAFILTRATION
PROCESS FOR PURIFICATION OF DYES USEFUL
IN FOODSTUFFS—.

In the Specification:

Page 1, line 5, after "1983" and before the period, insert —, now abandoned —.

In the Claims:

Claim 1, line 38, after "400 p.s.i.g.", insert —, at a pH from approximately 6.0 to 9.0, —.

REMARKS

The interview granted applicants' below-signed representative at the Patent and Trademark Office on May 9, 1985 by Examiner Higel is acknowledged. The foregoing amendments are believed to be in keeping with the understandings reached at the interview. It is believed that these amendments, taken together with the following remarks, will overcome all outstanding objections and rejections and place this application in condition for allowance.

The title has been amended herewith, in response to the Examiner's requirement, so as to be more specifically descriptive of the claimed invention.

The amendment in the specification at page 1, line 5 is made in order to provide the present status of parent application Serial No. 481,038.

The only other outstanding issues are the rejections of Claims 1-17, i.e. all the claims in the case, under 35 U.S.C. 103 as "being unpatentable over Booth et al. (U.S. Patent 4,189,380)" and as "being unpatentable over Colour Index in view of Booth et al.", the Colour Index reference being the citation in the Third Edition, Volume 4, at page 4087 of entry number 15985. These rejections are traversed for reasons which follow.

Booth discloses a process for purifying polymeric colorants by ultrafiltration through an appropriate semi-permeable membrane. The colorants purified by the process are dyes of the azo and anthraquinone classes, which are useful as non-absorbable colorants in edibles (column 2, lines 28-37). The dye chromophores are attached to a polymeric backbone, and the total polymeric molecules have a molecular weight greater than 1,000 Daltons. The Dalton is a unit of molecular weight used to express the molecular weights of extremely large molecules, such as polymers, one molecular weight unit being equal to 1.65×10^{-24} Daltons. Therefore the minimum molecular weight

of the polymeric dyes purified by the Booth et al. process is:

$$\frac{1,000 \text{ Daltons}}{1.65 \times 10^{-24} \text{ Daltons/M.W. unit}} = 0.61 \times 10^{27}$$

or 610×10^{24} ! In contrast, the dyes purified by the present process (in the form of their sodium salts) have molecular weights ranging from around 376 to 588:

FD and C Red 40	496.43
FD and C Yellow 6	451.37
Carmoisine	588.49
Amaranth	502.44
D and C Yellow 10	376.34

Thus the polymeric materials purified by the Booth et al. process have molecular weights differing from those purified by the process of the present invention by a factor of around 1×10^{24} . Obviously the process parameters required in the Booth et al. process for purification of such extraordinarily large molecules would, by necessity, be vastly different from the parameters required to purify the relatively small dyestuffs which are purified in the present process. Thus it is not believed that the Booth process could be fairly urged to even remotely contemplate purification of the relatively small molecules purified in accordance with the instant invention.

Moreover, for reasons that are not given by the patentee, the Booth et al. process requires the addition of salts to the feed solution (i.e. the "retentate"):

"It is the essence of the present invention to maintain above a certain level the concentration of salt in the retentate resulting from the ultrafiltration purification of a polymeric dye solution during at least two diavolumes of ultrafiltration, preferably during 3 to 20 diavolumes and more preferably during from 4 to 15 diavolumes. This can be effected either by loading the initial feed with an amount of salt sufficient to maintain the required minimum salt level

throughout the diafiltration or, and this is generally preferred, by maintaining the salt concentration by adding salt with the diafiltration makeup solution. A combination of these two methods may be used as well." (Underlining added) (Column 3, lines 33-46)

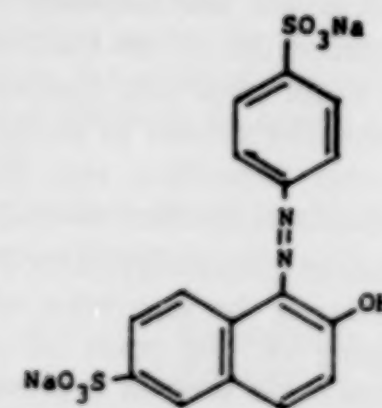
The present process does not require such addition, and in fact the process conditions are intended to remove salts, as expeditiously and as completely as possible, so that the product can be obtained at a level of purity far exceeding regulatory purity requirements. (See the tables at pages 16, 17, 20, 21, 23, 24 and 25 of the present application for a comparison of the specifications obtained by the purification of various dyes in accordance with the instant process with the specifications required by the FDA.)

Moreover, it is noted that Booth et al. state that it is "often of advantage to add and/or maintain additional materials to/in the ultrafiltration feed" (column 4, lines 35-36), such additional materials, including pyridine (column 4, line 39) or a base, being added in order to maintain the pH above 9, and preferably from 11 to 13 (column 4, lines 49-51). And finally, the ultrafiltration in the Booth process is carried out at pressures from 25 to 200 p.s.i.g., preferably at 50-150 p.s.i.g. and more preferably at 75-125 p.s.i.g. (column 2, lines 42-44). The process of the present invention, in contrast, is carried out at much higher pressures, i.e. 200-400 p.s.i.g. (specification, page 10, lines 7-9 and page 10, line 35 to page 11, line 8), and at much lower pH's, i.e. approximately 9 but preferably 6.0 to 8.0 (specification, page 12, lines 31-37).

In summary, it is submitted that the only feature the claimed process has in common with the Booth process is that both processes relate to the ultrafiltration purification of colorants used in foodstuffs. Otherwise, the proc-

ess disclosed by Booth et al. is submitted to be so totally dissimilar, in all its critical parameters, to the present process that the Booth process is in no way suggestive of the present invention. Nevertheless, in order to further highlight the process parameters of the instant process, and in accordance with the understanding reached at the interview, the pH range of 6.0 to 9.0 has been inserted in Claim 1 at line 38. Support for this amendment is found in the specification at page 12, lines 31-37.

The further rejection of Claims 1-17 for alleged unpatentability over Colour Index in view of Booth et al. is not understood. The Colour Index entry, reference number 15985 relied on by the Examiner, depicts the compound of the formula:



identified as "C.I. Food Yellow 3", which chemically is the disodium salt of 1-[(4-sulfophenyl)azo]-2-naphthol-6-sulfonic acid and which is thus identical with F D and C Yellow 6 (specification, page 4, lines 26-30), one of the species within the ambit of the present invention. Applicants, of course, acknowledge that F D and C Yellow 6, and all of the other four foodstuff dyes purified by the present process, are well known in the art. However, the here-claimed process is not founded on the structural novelty, or even the structural unobviousness, of the dye-stuffs which are so purified. Rather the patentability of

the instant process is submitted to rest on the novelty and the unobviousness of the process conditions, which are in no way taught or even suggested by Colour Index. The shortcomings in the Booth et al. reference have been discussed in detail above, and for reasons already given it is believed the present process is patentable over the combination of Colour Index with Booth et al.

The Examiner's attention is directed to several references which were not known to applicants at the time this application was filed and which may be pertinent to the question of patentability. Osei-Gyimah U.S. Patent 4,415,455 discloses semipermeable membranes and processes for their preparation for use in the desalination of water by ultrafiltration. The membranes operate by retaining salt species in the ultrafiltration feed water, and thus the retention properties of the membranes which are needed to remove small ionic salt species are vastly different from the properties needed to permit the passage of salt species, and other impurities, into the filtrate in the purification of dyestuffs as in the instant process.

Erzinger (assignor to Ciba-Geigy) U.S. Patent 4,452,608 addresses the problem of the separation of dyes into, and the resultant clogging of, the pores of an ultrafiltration semi-permeable membrane used in a process for the purification of dyes. The patentee overcomes the problem by adding a nonionic surfactant to the feed solution of the dye. The presence of surfactants, of course, would be undesirable in foodstuff dyes, and the Erzinger reference does not address the problems of the purification of such dyestuffs.

Horlacher et al. (assignors to Ciba-Geigy) U.S. Patent 4,466,900 discloses a process for preparing storage stable solutions of fluorescent brightener compositions by ultrafiltration of the brightener solutions through semi-permeable membranes containing ionizable groups. The membranes are typically made of cellulose acetate, polyacrylonitrile or copolymers of acrylonitrile and vinyl alcohol or vinyl

acetate (column 4, lines 2-6) and are modified in order to introduce the ionizable groups. The latter can be sulfato, sulfonic acid, sulfonic acid amide, carboxylic acid, carboxylic acid amide, hydroxyl, thiol, isocyanate or thioisocyanate, ammonium, phosphonium or sulfonium groups (column 4, lines 41-47). The rationale for the use of such ionizable groups on the ultrafiltration membrane for the purpose of obtaining storage stable aqueous solutions of the fluorescent brighteners is not given by the patentee, but in any case such groups are not essential to the purification of dyestuffs in accordance with the present invention, and the Horlacher process is not believed to be suggestive of the instant process.

The Bollenback et al., Adams et al., Teed et al., EPO, South Africa, Osmonics and Spatz I and II references, which the Examiner notes have been "cited by applicants, (and) show the state of the art" but not otherwise relied on are not believed to negate patentability of the subject invention.

This application is believed to be in condition for reconsideration and allowance, and such actions are respectfully solicited.

Respectfully submitted,
ROBERT W. J. REBHahn *et al.*

By /s/ William G. Webb
WILLIAM G. WEBB
Their Agent
Registry No. 19,861
Telephone (518) 445-8294

WGW:dd

May 14, 1985

Address:
B. Woodrow Wyatt
Sterling-Winthrop Research Institute
Rensselaer, New York 12144

[Record of Interview]

IN THE UNITED STATES PATENT AND
TRADEMARK OFFICE

Group Art Unit 124

Examiner: F. D. Higel

IN RE PATENT APPLICATION OF
ROBERT W. J. REBHahn *et al.*

Serial No. 677,118

Filed November 30, 1984

For: PROCESS FOR PURIFICATION OF DYES USEFUL IN
FOODSTUFFSRECORD OF INTERVIEWHon. Commissioner of Patents and Trademarks
Washington, D.C. 20231

Sir:

This paper is not an amendment, nor is it intended to be responsive to the outstanding Official Action (Paper No. 3) mailed February 14, 1985. Rather it is only intended to make of record the substance of an interview at the Patent and Trademark Office on May 9, 1985 between applicants' below-signed representative and Examiner Higel.

Concerning the Examiner's requirement to provide a more clearly descriptive title, applicants' representative proposed changing the title to read—ULTRAFILTRATION PROCESS FOR PURIFICATION OF DYES USEFUL IN FOODSTUFFS—, which the Examiner stated would meet his objection.

The discussion then turned to a consideration of the rejections of all the claims under 35 U.S.C. 103 as being unpatentable over Booth *et al.* U.S. Patent 4,189,380 alone or in combination with Colour Index, Third Edition, Volume 4, page 4087, entry number 15985 (1971). As to Booth, applicants' representative pointed out four major points of difference between the ultrafiltration process disclosed by Booth and the process here-claimed, namely (1) the enormous differences between the molecular weights of the dyes purified by Booth (i.e. >1,000 Daltons) and those purified by the present process (i.e. molecular weights from 376 to 588); (2) the requirement in the Booth process to add salts to the retentate; (3) the very high pH ranges deliberately sought in the Booth process, i.e. above 9.0 and preferably 11 to 13 by addition of basic materials to the retentate, in contrast to the relatively low pH's used in the present process (i.e. from around 6 to around 9), as disclosed in the instant specification at page 12, lines 31-37; and (4) the rather low pressures used in the Booth process, i.e. 25-200 p.s.i.g., preferably 50-150 p.s.i.g. and more preferably 75-125 p.s.i.g., as disclosed at column 2, lines 40-46 of Booth in contrast with the higher pressures required by the present process (i.e. 200-400 p.s.i.g.). The Examiner stated that, on presentation of such arguments, coupled with an amendment in Claim 1 inserting the pH range from 6 to 9, in accordance with the specification disclosure at page 12, lines 31-37, he would reconsider his rejection as based on Booth.

The rejection founded on Colour Index was apparently based on the Examiner's erroneous belief that certain dyes are being claimed in the instant application as products *per se*, and he readily agreed that the Colour Index reference would not support a rejection of the present claims directed to a purification process.

Applicants' representative handed the Examiner one copy each of U.S. Patents 4,415,455 (disclosing a process for ultrafiltration desalination of water); 4,452,608 (directed to overcoming the problem of the plugging of pores in membranes used in the ultrafiltration purification of dyes and the solution of the problem by the addition of a surfactant); and 4,466,900 (directed to the preparation of stable solutions of fluorescent dyes by ultrafiltration through membranes containing ionizable groups conjugated to the membrane). Applicants' representative indicated that he did not believe any of the references, alone or in combination, negated patentability of the present invention, but left the references with the Examiner for his own independent consideration.

Respectfully submitted,
REBHahn *et al.*

By /s/ William G. Webb
WILLIAM G. WEBB
Their Agent
Registry No. 19,861
Telephone: (518) 445-8294

WGW:dd

May 14, 1985

Address:

B. Woodrow Wyatt
Sterling-Winthrop Research Institute
Rensselaer, New York 12144

[Notice of Allowability]

UNITED STATES DEPARTMENT OF COMMERCE
Patent and Trademark Office

Address: Commissioner of Patents and Trademarks
Washington, D.C. 20231

Serial Number [677,118]

Filing Date [11/30/84]

First Named Applicant [Rebhahn, R.]

* * * *

Paper Number 5

Date Mailed: [06/26/85]

NOTICE OF ALLOWABILITY

PART I

1. ☒ This communication is responsive to applicants' amendment filed 05/17/85.
2. ☒ All the claims being allowable. PROSECUTION ON THE MERITS IS (OR REMAINS) CLOSED in this application. If not included herewith (or previously mailed), a Notice Of Allowance And Issue Fee Due or other appropriate communication will be sent in due course.
3. ☒ The allowed claims are 1 to 17.
4. ☐ The drawings filed on _____ are acceptable.
5. ☐ Acknowledgment is made of the claim for priority under 35 U.S.C. 119. The certified copy has [] been received. [] received. [] been filed in parent application Serial No. _____, filed on _____

6. ☐ Note the attached Examiner's Amendment.
7. ☐ Note the attached Examiner Interview Summary Record, PTOL-413.
8. ☐ Note the attached Examiner's Statement of Reasons for Allowance.
9. ☐ Note the attached NOTICE OF REFERENCES CITED, PTO-892.
10. ☐ Note the attached INFORMATION DISCLOSURE CITATION, PTO-1449.

* * * *

/s/ Floyd D. Higel
 FLOYD D. HIGEL
 Primary Patent Examiner
 Group Art Unit 124

DR. WAYNE COOK (DIRECT)

[192] Q. And then is it also a requirement of your patented process that the pH be such that it doesn't—or that it's compatible with the chemistry in the process itself?

A. Yes, that's right. And in the case of the lower limits, the issue there had to do with—well, we're going all the way back to the coupling reaction. As part of that reaction, sodium bicarbonate is generated as a by-product. I think I mentioned that a few minutes ago when we were talking about salts that are present here, probably as most of you are aware if you get sodium bicarbonate, it can be a very acidic situation. It foams. And carbon dioxide gas comes up and foams all [193] over the place.

What we found is that if you lower the pH of the coupling solution much below about six, there is tremendous foaming problems in the plant. And then like having something small foam up or so in your home, here you've got several thousand gallons of stuff that's foaming up going all over the floor and going all over the plant, so that's an undesirable situation.

Q. In terms of whether or not the process will work to separate the dye from the impurities, does it make a difference what pH you operate at?

A. Not to my knowledge. I would qualify that and say you probably would not go much below the pH of about two. And then you start getting into material problems.

Q. That would be in the equipment itself?

A. Yes.

Q. Why don't you put something on there in terms of the result you're trying to achieve, that is, separate the dye from the impurities, what effect the pH has?

A. It just says the pH has minimal effect on dye purification.

* * * *

[324] Q. All right. In the patented process in claim one, what's the purpose of the pH, generally?

[325] A. Generally speaking, the purpose of the pH is to maintain membrane stability and also to remain, more or less, in a neutral pH range for purposes of generating a, more or less, neutral product.

Q. Why do you want a, more or less, neutral product?

A. Because that's the product as specified by the FDA.

Q. Okay. Now when you say you want to maintain membrane stability by this pH, what do you mean?

A. For cellulose acetate membranes such as those used at the time that this invention was filed and recorded, those membranes are destroyed at certain pH's.

Q. All right, so you basically want to pick a pH that doesn't destroy your membrane?

A. That is correct.

Q. Now, this particular range that you have given in the patent is approximately six to nine. What did you have in mind when you used the word "approximately"?

MR. TAFT: Your Honor, again, I'll object to that. That was—

THE COURT: He may answer.

A. Well, certainly on the upper end of that range, we had an idea of how far we could go and still [326] retain the membrane integrity. On the lower end, the value of six was more a consequence of the process, and that we ended up there was not that particularly important that it be six.

Q. It was not particularly important?

A. No.

Q. All right. What pH is Warner-Jenkinson using in its process to make FD&C Red 40 dye?

A. At that particular stage in the process, my understanding is that their pH is about five.

Q. All right. And that would be—let me ask you this: At a pH of five, is Warner-Jenkinson performing

any function differently from the function that the pH's in the patent claim perform?

A. As far as the membrane separation process is concerned, no.

Q. All right. And does the pH have any—is a pH of five or six, whether it's five or six, have any difference, would it make any difference in the way the membrane operates, given the particular type of membrane that Warner-Jenkinson is using?

A. No, none that I know of.

Q. One of the differences that Warner-Jenkinson says exists between their process and the patented process is that there is an impurity called triazine in [327] the reaction mixture. Are you familiar with what a triazine is?

A. Yes.

Q. Where does the triazine in this reaction mixture come from?

A. It's a reaction by-product of the diazonium salt with paracresidine sulfonic acid.

Q. So that would be one of the by-products that you described before?

A. Yes.

Q. Is that something that the FDA likes to see in the by-product?

A. No.

Q. Now, Warner-Jenkinson also claims, in this case, that the reason they operate at a pH of five is to get rid of the triazine. Can you explain to the jury why that is?

A. The triazine impurity is unstable under acidic pH conditions, and so by operating at a lower pH, you can—or at least a specific pH, you can destroy the triazine.

Q. All right. So, a pH of five would be an acidic pH?

A. Yes.

Q. And according to the chart you made [328] yesterday, anything from zero to seven—or from zero to less than seven would be acidic?

A. That is correct.

Q. All right. Would a pH of six be considered acidic pH?

A. Yes, it would.

Q. Is triazine also destroyed at a pH of six?

A. Yes, it is.

Q. Now, Dr. Cook, I would like you to turn in your exhibit book, if you would, please, to Exhibits 30—we'll start with Exhibit 30.

MR. TAFT: Pardon me, Your Honor. May I go back over and check the exhibit books?

Q. All right, what is Exhibit 30, Dr. Cook?

A. Exhibit 30 is a copy of an article from the Journal of The Association of Official Analytical Chemists, Volume 67, Number four, 1984.

Q. All right. Who was this article written by?

A. Naomi Richfield Fratz, F-r-a-t-z.

Q. Who is that person with?

A. The Food and Drug Administration, Division of Color Technology.

Q. Is that the agency that's responsible for certifying food coloring?

A. That is correct.

[329] Q. Now, I would like you to look at Exhibit 31 and identify that for the jury, please.

A. This is a copy of another article from the same journal, The Association of Official Analytical Chemists. The particular article is from Volume 59, number three, 1976.

Q. Who is this article written by?

A. D. Douglas Fratz, F-r-a-t-z.

Q. And who is he with?

A. Also with the Division of Color Technology of the Food and Drug Administration.

Q. All right. What is Exhibit 32?

A. This is another article entitled—excuse me, I had to find the reference. This is another article from the Journal of The Association of Official Analytical Chemists, Volume 59, Number one, 1976.

Q. Who wrote this article?

A. John E. Bailey, B-a-i-l-e-y, and Elizabeth A. Cox, C-o-x.

Q. Who are they with?

A. They are also with the Division of Color Technology of the Food and Drug Administration.

Q. What is the conclusion reached in these three articles written by members of the Division of Color Technology of the Food and Drug Administration?

[330] A. The principal conclusion from these articles was that at pH's of around six, or acidic pH's, the triazine found in Red 40 decomposes.

Q. What do you mean, it decomposes?

A. It reverts to impurities, including cresidine sulfonic acids.

Q. That was one of the starting materials?

A. That is correct.

Q. So is there any difference, chemically, based upon your knowledge, as well as those articles from the Color Technology Division of the Food and Drug Administration, as far as the destruction of triazine is concerned, whether you operate at a pH of five, as in the Warner-Jenkinson process, or a pH of six, as in the patented process?

A. Not to my knowledge.

* * * *

[333] Q. All right. Based upon the statement of Dr. Solter's letter of May, 1986, that they wish to recover dyes from the feed solution of various FD&C food dyes of pH's of five to eight, what conclusion can you draw from that about the criticality of the pH, as far as the dyes are going to be subject to the membrane separation are concerned?

A. Well, I would infer from this that there is no major difference between pH five and pH eight or in [334] between.

* * * *

DR. WAYNE COOK (CROSS)

[385] Q. Yes. Now, we have membrane as a selection and that was pore size, pressure, that was 200 to 400?

A. To the best of my recollection, yes.

Q. And pH six to nine and pore size five to 15.

A. No, I don't think that was the correct pH.

Q. What's the correct pH range?

A. When we talked about the membrane stability, we talked about the membranes being stable at pH range from two to eight, that was specified in the Osmonics literature, and they asked us to provide solutions that fell within that range.

Q. So you want to make this two to eight?

A. Well, that was the pH range that they asked us to provide the solution at something that fell within that range, so we would not destroy their membranes for that test.

* * * *

[412] Q. So the membrane was selected by Osmonics, Osmonics told you what the pore size was; correct?

A. Well, the membranes were selected by Osmonics.

Q. Right. And then next we go to the 200 to 400 psig which is the pressure which was the Osmonics process; correct?

A. Those are conditions that work for this process, yes.

Q. As specified, as recommended by Osmonics; correct?

[413] A. They were initially suggested by Osmonics, yes.

Q. And then the pH of 6 to 9, which for the most part falls in the pH range of 2 to 8 which was recommended by Osmonics; correct?

A. That's correct.

* * * *

DR. RILEY KINMAN (DIRECT)

[489] Q. And what were you able to determine by reviewing that information?

A. The primary high pressure is higher than the Hilton Davis pressure. The intermediate pressure may or may not be very similar to the Hilton Davis primary pressure of 400 psi, pounds per square inch. The final pressure is usually in the range of 200 to 400 pounds per square inch pressure, which is in the Hilton Davis patent, is in the pressure range that they operate at.

Q. Why don't you pull up another board, if you would, and put those conclusions on it about what you determined the Warner-Jenkinson operating pressures to be?

A. Well, the primary pressure was usually around 500 plus or minus 20 or so. This is psi, pounds per square inch. The intermediate would vary as the process goes on, but, generally speaking, it is somewhere around 350 to 450 psi. And then the final would be less than 400 psi and it would be somewhere in the range of 200 to 400.

Q. All right. Let me put something up on the board. This is a claim chart, Plaintiff's Exhibit 7B, the second page of the claim. It calls for hydrostatic pressure of [490] approximately 200 to 400 psig. Based upon the review of Warner-Jenkinson's operating pressures, which pressures on which membranes lie within the range specified in the claim of the Hilton Davis patent?

A. Initially in their operating instructions they indicate that the pressure should be set at 500 plus or minus 20 psi. So this means the initial bank of membranes is seeing whatever that pressure setting was. Now, there were times in the operating log that it was down in the 400's, so the first bank of membranes is seeing that initial pressure.

Now, once you get inside the membrane modules, I can't say specifically which membrane is seeing exactly what pressure because this is both parallel and series op-

eration. So, the flow is moving all of the time through that bank of membranes on the concentrate side, because they are feeding back to the feed tank and so the pressure is downgrading from whatever that initial starting pressure is down to whatever that final pressure is that you read on the gauge. So a major portion of the membrane is actually receiving pressures in this 200 to 400 pounds per square inch gauge, the same as the claim in the patent.

* * * *

[494] A. * * * When we have a semipermeable membrane and we have water, this is H₂O on both sides, if there is a difference in concentration of these ions—I'm showing chlorides or sulfates over here and no chlorides and sulfates over here—then mother nature will try, with this water, to equalize the concentration. In other words, water in this case would pass over to dilute out this concentration of ions. So that when equilibrium is obtained, in other words, steady states, mother nature is happy, the concentration is the same on both sides of this semipermeable membrane.

Now, when the concentration is different, then there is what we call an osmotic pressure built up. In order to make that watering through there, you have to increase the pressure above osmotic pressure. And that's what we are doing in both the Hilton Davis process and in the Warner-Jenkinson process to prepare these dyes. We are putting sufficient pressure to overcome the osmotic pressure and push water through the [495] membrane plus impurities through the membrane with the water. In other words, not only can water flow through the membrane, but these ions can flow through that membrane also. In other words, these holes, passageways, if you will, are open to the fluid on the concentrate side and they are open to the fluid on the permeate side. And those ions can actually move back and forth through that water solution across that membrane, and we use the term it "transported" across the membrane.

Now, getting back to the actual process now. Since there will be an osmotic pressure caused by the dye and the various salts that are with the dye in the solution, and this combination of substances is greater than just clean water, which is on the permeate side, so there will be an osmotic pressure built up on this membrane and it will have some value. Now, that value will be a function of what's in here. In other words, you can't tell just by looking what the osmotic pressure is without making a measurement.

Now, the important thing is that this pressure, whatever it is, and let's just call it X, to make water flow through the membrane, we have to put a driving pressure using Warner-Jenkinson's term of primary pressure, if you will, that has to be greater than this X. Now, once you get the pressure greater than, greater than the X, water will move through the membrane and you would be able to see permeate flow indicated on your flow recording gauge, so many gallons per minute of [496] water permeate coming through the membrane.

Q. Where would that permeate gauge be located in the Warner-Jenkinson system?

A. The permeate flow would be coming out here.

Q. Is that where the permeate gauge would be located?

A. Yes.

Q. Go ahead.

A. Now, as I indicated to you early on, there is no reason for increasing the pressure greater than in achieving the osmotic pressure plus some driving force to push the permeate through the membrane and we try to keep that pressure as low as possible.

Now, let me give you an illustration. In the early days of membrane work, we were trying to use membranes for desalting sea water and because of the high concentration of salts in sea water, the pressure required to make clean water from sea water or desalt it was 15,000 psi, pounds per square inch. And this is very high pressure.

And it required a very high pressure pump and it required very high pressure valves and fittings, and all of these things.

Now, with the new membranes, we are able to use low pressure, which is a much less demand as required for energy. So we are down in the operating range of these two companies' processes of the 200 to 400 pounds per square inch, and this has been one reason why the ultra-filtration RO process has been [497] more widely used now. We are able to get those pressures down in a more reasonable range.

Q. Is there any difference in the way that the membrane and the Warner-Jenkinson process is functioning at the pressures it is seeing in comparison to the way that the membrane in the claims of the Hilton Davis patent is operating under a pressure of 200 to 400 psi?

A. It is doing essentially the same thing.

Q. Explain what it's doing and how it is doing it.

A. Essentially, and let's use this side as being the pressure side, we are putting a pressure greater than osmotic and by doing that we are pushing water plus impurities through the membrane and we are achieving the desired result of a purification of the dye solution. This is water plus dye and a removal of impurities from this water plus dye solution and, of course, a concentration, then, of the final end product.

Q. Is that result being achieved in both the Warner-Jenkinson process and in the process that's described in the claims of the patent?

A. Yes.

Q. Now, are you aware in this case of any different results that Warner-Jenkinson is claiming it achieves with its process which compared to the results achieved with the process in the patent as far as the pressure is concerned?

A. Well, they indicate some reason that they are using [498] the higher pressures, but I do not see any basic difference in the process.

Q. Is one of the reasons they indicate that they get less dye loss?

A. Yes.

Q. And what has your investigation shown, if any, as to whether or not Warner-Jenkinson is achieving less dye loss at their higher pressures than Hilton Davis achieves?

A. I do not see any real change in the dye loss between the two processes.

Q. Could you expand on that, explain that a little bit more?

A. Well—

Q. What exactly did you look at that to come to that conclusion?

A. I looked at their process out there, and at the higher pressures they appeared to be losing more of their dye solution around the pump packing and casing and so forth. Then, in looking at the actual processing sheets, the actual concentrations of impurities, and the conductivities and so forth on the final solution are equivalent to the Hilton Davis final solution.

* * * *

[512] Q. One other requirement in this claim is that the process operate at a pH approximately 6 to 9. While you were at Warner-Jenkinson and inspecting their process equipment, did you observe any instruments on their dye processing equipment which were used to record pH?

A. Yes.

Q. What did you observe?

A. Well, I observed on the Red Dye 40 Osmonics unit that was treating the dye pH of 6.

Q. How did you observe that?

A. By reading the strip chart recorder that they had on the unit.

Q. What's a strip chart recorder?

A. A strip chart recorder is a roll of paper that has points indicated. In this case, we are measuring pH and there is an actual line drawn at the particular number

that we talked about here. In this case, it was a pH of 6, and there is a continuous line on there over time as to what the pH is. If the pH changes, the needle would move up and down on that chart. But I did observe the pH of 6 that day when we were there.

* * * *

DR. RILEY KINMAN (CROSS)

[520] Q. Now, I believe you just told these folks that, with regard to the pH of 6, that you observed all of our machines and you observed on one of them on the pressure scale that it says a pH of 6. Didn't you tell these people that?

A. I observed it on the Red Dye 40 machine that there was a pH of 6, yes.

Q. But what you didn't tell these people is that that machine was not running at the time, was it?

A. The machine that I observed was running.

Q. I'm talking about the one with the pH of 6. Are you saying that machine was running at the time?

A. There were four different machines that we observed, and, as I recall, three out of the four were running.

Q. And one was shut down for cleaning, wasn't it?

A. Could have been.

Q. And that was the one, wasn't it?

A. Could have been.

* * * *

DR. RILEY KINMAN (REDIRECT)

[552] Q. At any of those upstream sides of the membrane, did you find the pressure to be actually within the range of 200 to 400?

A. Yes.

* * * *

DR. ROBERT KESTING (DIRECT)

[753] Q. Can you tell the jury generally what the pH scale is and what it means?

A. Yes, I can. I would just like to preface this by saying people in general, and scientists in particular, like to work with small numbers. Just easier to grasp, easier to remember, easier to understand. So they will convert complicated numbers into small, frequently whole numbers. And that's what was done in the case of, for example, the Richter scale for earthquake damage or earthquake magnitude, and rather than speak of numbers like a thousand and ten thousand and a hundred thousand, they speak of Richter scale, for example, of typically between 1 and, say, 9. And those units differ from one another in magnitude. In other words, going from 1 to 2 was not a small step. It's a large step. It's a ten-fold increase in magnitude as shown on the seismograph scales. It [754] can be actually a 31-fold increase in the energy that's involved in each step.

In other words, an earthquake with a Richter scale number of 7 is not simply somewhat removed from an earthquake of 8. There is big difference in there. It is a ten-fold difference in magnitude and a 31-fold difference in the energy. So it is easier to talk about 8 than to say 230. And that's the reason why they use these scales that go back to some of the fundamentals and much easier to talk about.

Same was done in the case of pH. pH is a way of describing the hydrogen ion concentration and the actual definition of—it is the law of the reciprocal. That's one over that particular hydrogen ion concentration expressed in moles per liter. So if you have one over ten to the minus two, that's a hundredth of a mole of hydrogen ions per liter. That would be a pH of 2. The difference between a pH of 2 and a pH of 3 is considerable. The pH of 2 is ten-fold higher concentration of hydrogen ions than a pH of 3. And so I'm just going through that to show you that these numbers, although they sound like they are

close together, they are not that close together in terms of actual concentrations. So I want you to be aware that the difference of somebody operating in a pH of 5 can be quite different than somebody operating in a pH of 6.

Q. Let's try and simplify this, and let me just use the [755] back of this board here. If we take the chart here, and we are going to have pH of 7 here, which is neutral. And then 6 and 5 and 4, and then you'll go 8, 9 in the other direction?

A. Yes.

Q. What are we going to put on this side?

A. That would be the hydrogen ion concentration.

Q. Could acidity be another term?

A. I would just be H plus, in parentheses, or in brackets.

Q. So now if we go down from 7 to 6, you say that's ten-fold?

A. Ten-fold increase in hydrogen ion concentration.

Q. That would be ten.

A. Right.

Q. But if we go down from 6 to 5, what happens?

A. Well, starting comparing that to 7, you would have another, it would be 100.

Q. So that would be 100-fold difference.

A. And you go in the other direction, you are going down, pH of 8 would be one hundredth.

* * * *

DR. ROBERT KESTING (CROSS)

[851] Q. I changed it. Warner-Jenkinson is running their process and it's running making this dye at a pH of 5. And I come up with a big bucket of something. And I dump that bucket of something into their vat and the pH goes up to 6. Does the machine quit?

A. No.

Q. Does it quit making the red dye, does the red dye quit coming out?

A. No.

Q. Let's turn it around. Let's go over to Hilton Davis. Go up to their machine. They are making red dye and yellow dye at a pH, say between 6 and 9. And now I take a big bucket of acid and I dump it in there and pH goes down to 5. Does the Hilton Davis process quit?

[852] A. I wouldn't imagine so.

* * * *

JAMES NOONAN (DIRECT)

[888] Q. Now, let me ask you, before you go into talking about that, where was Warner-Jenkinson in October 1986 when you first learned about this patent, where was Warner-Jenkinson in terms of its progress in its own ultrafiltration processes of Red 40 and Yellow 6 dyes?

A. We had behind us about four years of doing research into the process, and, in fact, we started, in about in 1982, we started researching the ultrafiltration process and had made a lot of progress. We already had the process pretty much worked out. We had some of the final touches to make and had worked already with a good number of the colors that we were selling and had actually produced as much as five thousand pounds of one of the colors that produced it using the ultrafiltration process that we had developed.

Q. And at the time that you came across this patent, what was left to do to totally convert over all of your manufacturing at that time to finish that, what was left to do?

A. Principally, it was ordering the equipment. We had rented equipment from three different suppliers of this equipment and we had done a lot of work with each—with all of the equipment, and we were at the point where—and it was all rented equipment up to that point. And we were just about ready to make the decision as to which of the products—the [889] equipment that we would purchase.

Q. You mentioned that when you found the patent you contacted your outside patent counsel, correct?

A. That's correct.

Q. And who was that or is that?

A. Mr. Donald Leavitt.

Q. And would you just tell us what you did and what happened then with patent counsel? Did you send the patent to him?

A. Pardon?

Q. Did you send the patent to him?

A. Yes, we did. We immediately obtained the patent and immediately contacted Mr. Leavitt, with whom we had worked for many, many years. He served our company in this area a long time. We had a lot of faith in him. He had a very good reputation. And so we immediately contacted Mr. Leavitt and he started the work that he had to do to get the information, get the patent file, so that he could familiarize himself with the situation so that we could see what action we were going to take.

Q. And what happened next?

A. Well, after we obtained the—after Mr. Leavitt obtained the history of the patent negotiations, we met and discussed what was the appropriate action for us to take based on his evaluation of what had been considered by the patent [890] office.

Q. Now, let me just stop you for a minute. Before you tell us about that meeting, would you take a look, please, at Defendant's Exhibit 589?

A. What number was that?

Q. 589, please. I just want to run through some of these exhibits as we go along just so we have into evidence to document what you did. Do you have that in front of you, 589?

A. Yes.

Q. Is that the letter that you sent to Mr. Leavitt, your outside counsel, sending a copy of the patent?

A. Yes.

Q. And that's dated November 6, 1986?

A. That's correct.

Q. Now, let me ask you this. Why did you send the patent to outside counsel? Why did you contact outside counsel? It might be obvious, but tell us why.

A. Well, it was very important because we were—we had done an awful lot of work in this area, as I have mentioned, over a four-year period. I don't know that we were necessarily considering patent in the process. We had done a lot of work on it, but a lot of it seemed to us to be just prior art, and so we felt that we should take a close look at this and see just exactly what our next step would be.

Q. So you sent the letter to the lawyer, you got some [891] other information for your counsel, Mr. Leavitt, and then I believe you said you had a meeting?

A. Yes.

Q. When was that?

A. We met on January 19, 1987, and in this meeting we had myself and some of our technical people who were involved in it, Dr. Solter and Dr. Bischoff.

Q. And Dr. Solter is sitting here at counsel table?

A. Yes, he is.

Q. And Mr. Leavitt, your patent counsel, is sitting back there in the pew to the right?

A. That's correct.

Q. So what happened at that meeting?

A. We discussed the patent, of course, and discussed what we had done in this area and we came to the conclusion that we felt that we were not infringing.

(Pause in proceedings.)

Q. What did you—you were starting to say that we discussed it and we came to the conclusion. Go ahead.

A. And that we came to the conclusion that what we were doing was the process that we had almost completely developed, we weren't quite finished with it yet, but it wasn't infringing on the patent.

Q. And why was that?

A. Because we were operating under a considerably [892] different operative method.

Q. Are you talking about your operating pressures up in the upper 400s, lower 500s?

A. That's correct.

Q. And your pH down at 5?

A. That's correct.

Q. And what was the advice to you at that time from your patent counsel, Mr. Leavitt, sitting back there?

A. Our advice was—his advice was to continue what we were doing and to keep him informed and that to just finish up on our process and then we would discuss the situation.

Q. Did he tell you about anything about your purchase of equipment and when you should get back to him for a formal written opinion?

A. Well, that was when we got to the point where we were at that stage that we certainly should get his opinion at that point in time. Yes, that was kind of certainly mutually understood, because we were not interested in just blatantly violating a patent.

Q. Now, I believe you have told us so far that he told you that the way you did your own process you didn't infringe, correct?

A. That's correct.

Q. Did he tell you anything about what he thought about the validity of the patent in the first place?

[893] A. Yes, he considered that the patent was not valid.

* * * *

[902] Q. And did he, in fact, give you a formal opinion letter?

A. Yes. On October 12 he submitted to us a formal opinion letter.

Q. And without reading that whole letter, did he tell you that as long as you operated above 450 and at a pH of 5.5 or below you wouldn't infringe that patent?

A. That's right. In addition to that, he felt that the patent was really not valid, which was a very important input.

Q. But you wanted to avoid all this anyway, so you wanted to also—you didn't want to infringe it if you didn't have to?

A. That's correct.

* * * *

JAMES NOONAN (CROSS)

[905] Q. Mr. Noonan, you testified a few moments ago that when you first learned about the Hilton Davis patent you were immediately concerned; that was your testimony, wasn't it?

A. Yes.

Q. And the reason that you were concerned, sir, was because Warner-Jenkinson had been working on the same sort of a project, correct?

A. Yes, that's correct.

Q. In fact, sir, your concern was specifically that there had been a patent issued on a process that Warner-Jenkinson had been working on, isn't that correct?

A. Yes, in general.

Q. But when you learned about the patent, you didn't [906] stop using the process, did you?

A. No, we didn't.

Q. In fact, at the time that you learned about the patent in October of 1986, Warner-Jenkinson was already making Red 40 in full batch loads and selling it, weren't you?

A. That's correct.

Q. And you said in your testimony that you were making—that you made about five thousand pounds up to that point, but, Mr. Noonan, I'd like to direct your attention to Plaintiff's Exhibit 90, which is a listing of

how much Red 40 Warner-Jenkinson actually made in 1986, and that was ten thousand pounds. Are you aware of that?

A. No. I had not seen these figures before.

Q. And in 1987 Warner-Jenkinson actually made 95,756 pounds?

A. In 1987?

Q. In 1987. Are you aware of that?

A. I'm not aware of all these figures.

Q. And during all this period of time, all the way up until October 12, 1987, a year after you first learned of the work of the Hilton Davis patent, you didn't have any opinions from your counsel at all, did you?

A. Yes, we did. We had an unofficial verbal opinion in our January meeting in 1987.

* * * *

[915] Q. And it is a fact, is it not, that from October 1986, when you first found out about the Hilton Davis patent, until October of 1987, you did not have a written opinion from Mr. Leavitt?

A. That's correct.

* * * *

DONALD LEAVITT (DIRECT)

[924] A. Yes. I have represented Warner-Jenkinson for over 25 years and in that connection have become familiar with the color industry and the dye industry.

Q. Let me ask you, Mr. Leavitt, when did you become a patent lawyer?

A. I was admitted to the bar in Illinois in Missouri in 1952 and became registered to practice before the United States [925] Patent and Trademark Office, I believe, in 1953.

Q. And have you been in like a law firm type private practice before the patent office, ever since then?

A. Yes, I have been in private practice since 1952.

Q. As a patent lawyer?

A. As a patent lawyer with the firm I am with now, yes, for 40 years.

* * * *

Q. When did you first become aware of this patent?

A. I first became aware of the Hilton Davis patent in November 1986.

Q. Now, how was that patent transmitted to you?

A. Mr. Noonan, the then president of Warner-Jenkinson, wrote to me on November the 6th, 1986 and transmitted a copy of the Hilton Davis patent. He had previously discussed this with [926] me over the telephone and this letter supplemented our telephone discussion, and he sent me a copy of the patent, which is the first time I ever saw the patent.

* * * *

[927] A. After I received Mr. Noonan's letter, I reviewed the patent itself to make certain that I had an understanding of what was described and claimed in the patent. I called Mr. Noonan on the 21st, I believe, of November, 1986, and told him that I had reviewed the patent and I suggested that we order a copy of the file history of the patent. That's a copy of the proceedings between the applicant and the patent office resulting in the issuance of the patent. I thereupon ordered a copy of the file history from my office on November 21, 1986. I don't have a record of when that was received by our office.

[928] Q. When you say a copy of the patent file, you mean you wrote to the patent office and you actually obtained the whole file from the patent office, not just the patent?

A. That's correct.

Q. Go ahead. You were saying you couldn't tell from your file or you don't have a letter telling exactly when you received it?

A. That was received probably sometime in December or January. I then reviewed the file history from begin-

ning to end to make certain that I was familiar with the entire proceeding of the application before it matured into the issued patent. We then arranged to meet with Mr. Noonan and others at Warner-Jenkinson on January 19, 1987. At that time, I completed—

Q. Pardon me one minute. I'm sorry. I shouldn't have interrupted. Go ahead.

A. At that time, I had completed my study of the patent itself and my study of the file history and all of the references that were cited in the prosecution of the patent application before the patent office. I met with Mr. Noonan, Dr. Solter, and Mr. Bischoff of Warner-Jenkinson on January 19, 1987.

* * * *

[929] A. At that meeting I was advised that Warner-Jenkinson had been working themselves on an ultrafiltration process since 1982. I was further advised that they had already been making pilot plant quantities of certain food colors by ultrafiltration, namely, FD&C Red 40, FD&C Blue 1, FD&C Red 3, FD&C Green 3, and I believe others. I was advised that, in the course of their own work and their work with the suppliers, the suppliers being Osmonics, Carre, and Niro, the three that I remember, that they had used various conditions, but at that point in time had settled on using pressures in excess of 450 pounds per square inch on up to pressures of 525 pounds per square inch. They also informed me that they were using pHs below 5.5, all the way down to 5.

[930] During that discussion, based on my study of the patent and the file history, I advised Warner-Jenkinson at that meeting that, using those conditions, they were not infringing the Hilton Davis patent because those conditions were outside of the scope of the claims of the patent.

I further advised them, I believe, that there was not infringement either on a literal basis, or under the so-called doctrine of equivalents. They told me that they were continuing to work on this and had not settled yet on the particular equipment that they would purchase for use on a commercial scale, but that they were continuing

their work and that it was likely that sometime during 1987 that decision would be made.

I suggested that when they did make the final decision, that they should inform me and then tell me what the exact operating conditions were, the exact process parameters, so that I could then render a formal written opinion on the matter based on what they were going to do commercially, not based on what they were experimenting with. And I left with the idea that they would keep me informed, and when they arrived at the point where the commercial equipment was settled upon and the process conditions to be used commercially were settled on, they would let me know and I would then render a formal written opinion.

Q. Now, you mentioned that you advised them that they [931] did not infringe the patent. Would you describe to these folks why, as of that meeting, January 1987, you had determined as patent counsel that what Warner-Jenkinson was doing would not infringe upon those claims in that patent?

A. The claims of the patent themselves are limited to an ultrafiltration process operated at hydrostatic pressure of 200 to 400 pounds per square inch. Also limited to operating at a pH of 6 to 9. The file history of the prosecution of the application, which resulted in the issuance of the patent, shows that the allowance of the claims by the examiner was predicated on the limitation in the claims to that pH range of 6 to 9 and the argument presented by the applicant's agent or attorney that the pressure range of 200 to 400 distinguished over the Booth et al. patent, which you have heard about in this courtroom.

Indeed, in one part of the file history, it was emphasized that the novel and unobvious features of the Hilton Davis invention rest on the process conditions, referring to the particular pH range that I have mentioned and the particular pressure range that I have mentioned.

Taking all that into account, we, first of all, note that a pressure of 450 up to 525 is clearly outside the range

of 200 to 400. A pH of 5.5 down to 5 is clearly outside the range of 6 to 9. Moreover, under the so-called doctrine of equivalents, we have to take into account, a patent lawyer has [932] to take into account another doctrine, the doctrine of wrapper estoppel. Under that doctrine, if an applicant argues—

* * * *

[933] Q. Rather than refer to the specific legal terms of doctrine of equivalents or doctrine of wrapper estoppel, rather than actually referring to those, what we would ask you to do is to talk about what you saw in the file and what they argued to the patent examiner and why that influence, how that is that that affects your opinion. Can you do that? I think where you were was you were talking about—you were talking about estoppel. If you would just pick up there and explain.

A. Well, having obtained the allowance of claims, only after amending the claims to insert the pH range of 6 to 9, and only after arguing that the pressure range of 200 to 400 is, quote, "much higher than the Booth range of 25 to 200," and I don't quite understand how 200 can be much higher than 200, but nevertheless, after those arguments and amendments were made, my opinion is that the claims cannot be stretched under the doctrine of equivalents to encompass numbers which are outside the 200 to 400 range and the 5—I'm sorry, the 6 to 9 pH range.

Q. Now, did you also notice, when you were going through the patent office's actual complete file—and which has been marked, by the way, as Defendant's Exhibit in this lawsuit—did you notice what the patent examiner determined as to whether the idea of applying ultrafiltration to these particular dyes, whether that the idea itself was something [934] that was patentable?

A. No. The examiner said that that idea was not new and he cited the Booth patent, and of course he distinguished over the Booth patent strictly on the basis

of a different pressure range and a different pH range, as you have heard discussed before.

* * * *

DONALD LEAVITT (CROSS)

[964] Q. Did you ever actually go out to Warner-Jenkinson and look at a real honest-to-goodness operating system to see what it was doing?

A. No, I did not.

Q. Mr. Leavitt, I'd like to place here on the easel, if I may, a diagram we've made earlier in this case by Professor Kinman, and he was describing what he saw when he went to Warner-Jenkinson as far as what their system looked like. And he drew the membrane modules here and he explained to us that he had actually observed from the system itself and from documentation what the pressures were on each one of these modules. You didn't do that, did you?

A. I asked the client what pressures they were using in terms of what the patent described as pressures. The pressure in the upstream side of the membrane which is the sense in which pressure is used in the patent.

Q. Did you understand that there is actually several upstream sides of the membrane? There's one here and there's also one here?

A. Possibly.

[965] Q. Did Dr. Solter explain to you that the initial pressure might be 525 but that the intermediate and the final pressures might be much lower than that?

A. No, we didn't discuss that.

Q. And Dr. Solter also didn't explain to you, did he, that in fact in the actual system that Warner-Jenkinson is using to practice this process the intermediate and the final pressures may be as low as in the 200 to 400 pounds per square inch range?

A. No.

* * * *

[969] Q. And under the column or the heading "pH," under "patent" Mr. Noonan says 6 to 9, and under the Warner-Jenkinson column he says 4 to 8?

A. Yes.

Q. And it was your testimony this morning, was it not, sir, that you were being informed or you said that you were being informed that their pH was really 5.5?

A. I was informed that they were actually operating at a pH of 5.4 or below.

Q. And in this letter a month before you rendered your formal opinion Dr. Solter is telling you that, "The parameters specified in the patent claims and those in our process are as follows," and he tells you the pH is 4 to 8?

A. I found out they weren't using a pH above 5.5.

Q. That is based upon what Dr. Solter told you?

A. Dr. Solter and others.

* * * *

DR. LANCE SOLTER (DIRECT)

[1004] Q. Okay. How long have you been working with dyes out at Warner-Jenkinson?

A. Well, I would start with the very first year in 1974, so I've been associated with dye chemistry for about 18 years.

Q. What I'd like to do is just take you right to what we're talking about in this case, and that is the ultra-filtration processes for your dyes.

Now, and if we can, what number is this?

MR. TAFT: Your Honor, I would like to identify—rather than get a sticker, I just wrote it on there, Defendant's Exhibit 632.

* * * *

[1005] Q. I wonder if you would first of all tell these folks how this exhibit was prepared?

A. Well, basically what we did was we went back through the documents and I listed the documents basically in order with the dates, and we reviewed them together and that's what this is.

Q. This is a time line, okay. And what does it represent?

A. Well, it represents basically in condensed form what we've done on ultrafiltration starting with 1982 and going through to the date that we discovered about the Hilton Davis patent.

* * * *

[1011] Q. Okay. Now, why did you want to take out triazene at the pH of 5 before you ever sent it up there in August of 1982?

A. Because that was a part of our normal process before filtration.

* * * *

[1040] Q. By the way, at this point in time you have already ordered one set of equipment, you are testing on another set, you have done this with your dyes that you have done. Did you [1041] have any idea that Hilton Davis had been up to Osmonics?

A. No, I did not.

Q. And this is January, the patent issued on December 24, 1985. I would just write that on there, 1985. Did you have any idea whatsoever, when you are issuing orders to rent equipment and now you are testing a second set of equipment, did you have any idea whatsoever that any patent had been issued to Osmonics?

A. No, I had not.

Q. I'm sorry, to Hilton Davis?

A. No, I had not.

* * * *

[1044] Q. And then we get into August and you are discussing with Osmonics to start up the larger rental unit at Warner-Jenkinson, and then by September you are producing full five thousand pound batches of Red 40 on Osmonics larger?

[1045] A. That's correct.

Q. And that's larger than—well, you do much larger ones now, don't you?

A. Yes, we do.

Q. And then, in August, you actually sold Red 40 from ultrafiltration to customers and you started producing * * * on the larger equipment?

A. That's correct.

Q. And those are full batch sizes?

A. That's correct.

Q. And then you discovered the Hilton Davis patent?

A. Right. Dr. Sujeeth did a literature search, and around October 14th, that's the notation he has in that book, he discovered the Hilton Davis patent.

Q. Now, up to this point in October that Sujeeth discovered the Hillton Davis patent, to your knowledge, did you, or anyone else at Warner-Jenkinson, have any knowledge whatsoever that Hilton Davis was doing ultrafiltration, that they had been to Osmonics or that they had any patent?

A. No, there was no such knowledge.

Q. None?

A. None.

* * * *

[1046] Q. All these tests were run before you ever saw the patent or ever had any idea that Hilton Davis was doing ultrafiltration, is that correct?

A. That is correct.

* * * *

[1047] Q. And so you had established process conditions at 450, 470, 500, 520, 520, on three different sets of equipment [1048] prior to ever hearing of the patent?

A. That's correct.

* * * *

[1055] Q. I'd like to talk about, then, once the patent was delivered to you, what happened after that?

A. After the patent was found, I don't remember if I called Jim Noonan or if I had Sujeeth to copy the patent to him or what, but I know that I did let Jim Noonan know about the patent immediately.

[1056] Q. What happened after that?

A. Well, after that, Jim sent a letter to Don Leavitt. I believe it was in November.

Q. Asking for a legal opinion?

A. Asking for a legal opinion, yes.

Q. And then after that, did you have a meeting with Mr. Leavitt and others and were you given a legal opinion verbally as to whether your process conditions—when I say “yours” I mean Warner-Jenkinson's process conditions—violated the Hilton Davis patent and whether the Hilton Davis patent was valid?

A. I am positive we were given a legal opinion before we kept on producing, but at that time we had already produced color before we learned about the patent. I don't remember too much about that meeting, quite frankly. I think, based on the notes that Don Leavitt has put in the testimony here, that I believe was January, was it?

Q. Yeah.

A. Yeah, but I do know for a fact that before I started production again I had a legal opinion.

Q. When did you start production again?

A. It would have to be after that January.

Q. So sometime either in January or before then you received some verbal legal opinion that you didn't infringe and the patent was probably invalid?

[1057] A. Yes.

* * * *

[1058] Q. And let me ask you about your process conditions. You have already told us what they were. When you actually then went forward and bought your equipment in late '87 and then in '88, did you change those process conditions, did you ever change them to operate differently than the way you had developed those process conditions before you ever knew about the patent?

A. No, we always ran those processes just like we told Don Leavitt. We were always above 450 psig and we were always below a pH of 5.5. In the case of pH, we

were always at 5. In the case of the pressure, we were between 480, 470 and 500 [1059] always.

Q. Why did you operate at the higher pressures?

A. We operated at the higher pressures because, as you would see, we got greater flux. This is more permeate going through the membrane at the same time. This means you got more removal of sulfates and of the intermediates, plus you would be able to concentrate faster at the higher pressures. Once you stopped the dye filtration, once the purification was done, then all you had to do was to concentrate the higher pressures. You would concentrate faster. We also found out that the dye, removal of the dye loss was less at the higher pressures.

Q. And did you find that out before you ever heard of this patent?

A. We knew that before we heard of the patent in October, yes.

Q. Now, when you then went forward and bought the equipment, the permanent equipment and actually started converting over all of your manufacture of these materials to the new equipment to ultrafiltration, did you, or, to your knowledge, any other person anywhere at Warner-Jenkinson, ever use anything from the Hilton Davis patent?

A. We never used anything from the Hilton Davis patent.

Q. Why not?

A. Well, we felt like, you know, first of all, we had a better process. That would be one reason. Number two, they [1060] had a patent. If that patent was valid, we could not use their process, but we did not need to. We had a better process and we had a legal opinion that said we were not infringing.

* * * *

Q. How long have you been operating at a pH of 5 to destroy triazene?

A. That goes back beyond 1982.

Q. And you have always done it that way?

A. Yes.

* * * *

GERARD GACH (CROSS)

[1262] Q. I want you to look through there, and I want you to pull out the paper that looks like this. It's got Warner-Jenkinson at the top and then it says "Ideas for second application test."

A. Um-hum. I have that.

Q. And what that paper says is "Warner-Jenkinson, ideas for second application test, red dye, dilute 10 percent solution to 5 percent like Hilton Davis."

A. That's correct.

Q. Then it says, "Use HD," Hilton Davis, "flow rate and dye passage figures for the 50 HCA." That's what it says in there, doesn't it?

[1263] A. That's right.

Q. And that's in contemplation of Warner-Jenkinson having done a second test on the Red 40 dye, wasn't it?

A. That's correct.

* * * *

DR. WAYNE COOK (CROSS)

[1578] Q. And I'm not talking about the chemistry, I'm talking about the process of ultrafiltration. Does the pH make a difference?

A. Between a pH of 2 and 8, I would not expect to see much difference.

Q. Okay.

A. For Red 40.

Q. Can we write that down, then?

A. Sure.

Q. So between pH of 2 to 8 you do not expect a difference?

A. As far as the membrane performance is concerned presuming that there is no chemistry changes. That's critical.

[1579] Q. We're going to get to that. I'm talking about whether or not—

A. I just want to make sure.

Q. —whether or not it's functioning to separate.

A. We're assuming that the chemistry does not change between the pH of 2 and a pH of 8, that all that's happening is that you're taking the same solution, we've changed the pH, and there's been no other changes in chemistry—

Q. Correct.

A. —will the membrane perform the same across that range. Is that—do I understand the question correctly?

Q. Yes.

A. Okay. Yes. I would not expect to see any major differences.

Q. Do not expect—

A. Major differences.

Q. —major differences. The reason I asked you that question is because Mr. Schmit asked you something similar at Page 193 of the transcript. The question was, "In terms of whether or not the process will work to separate the dye from the impurities, does it make a difference what pH you operate at?" And your answer was, "Not to my knowledge." Are you consistent?

[1580] A. I think so.

Q. It doesn't make a difference.

* * * *

[1581] Q. And the reason you chose 9 was because that's just where the coupling reaction pH lies?

A. That's about where it ends up, yes.

Q. And so you put that in your patent application?

A. Yes.

Q. And then even though Osmonics had told you you could operate 2 to 8, you cut it off at 6?

A. Right.

Q. And the reason you did that was because of your Red 40 process would foam if you went below 6?

A. That's correct.

* * * *

DR. WAYNE COOK (REDIRECT)

[1623] Q. During the time that you were running all these tests, going all the way down to 2.2 and 3 and 4 and 5, was the process running the test actually doing what it was supposed to do?

A. Yes.

* * * *

Defendant's Exhibit 599

LAW OFFICES OF

SENNTGER, POWERS, LEAVITT AND ROEDEL

611 Olive Street
St. Louis, Mo. 63101

October 12, 1987

Mr. James E. Noonan
Warner-Jenkinson Company
P.O. Box 14538
2526 Baldwin Street
St. Louis, Missouri 63178-4538

Dear Jim:

Confidential
Attorney-Client Privileged
File 7285

This letter and opinion will confirm and supplement our recent conference regarding Hilton-Davis U.S. patent no. 4,560,746 directed to an ultrafiltration process for the purification of dyes useful in foodstuffs. As background for the opinion expressed herein, we have studied the Hilton-Davis patent, the file histories of the issued patent and of the abandoned original application Serial No. 481,038, and the prior art of which we are currently aware. We have also taken into account the information which you and your technical people have provided to me during our various meetings on this subject.

Possible Infringement of the Hilton-Davis Patent

The claims of the Hilton-Davis patent are directed to an improvement in a process for the purification of a dye from among those enumerated in claim 1 of the patent, the improvement comprising the steps of subjecting an aqueous solution of the reaction mixture resulting from the recited coupling or sulfonation reactions to ultrafiltration through a membrane having a nominal pore diameter

of 5-15 Angstroms under a hydrostatic pressure of approximately 200 to 400 p.s.i.g. at a pH from approximately 6.0 to 9.0 to cause separation of impurities from the dye, and, when substantially all of the impurities have been removed from the concentrate containing the dye, recovering the dye in approximately 90% purity from the concentrate by evaporation of the concentrate to dryness. Claim 1 defines the improvement in the above-stated terms while dependent claims 2-17 define other features of the alleged invention, e.g. claim 2 specifies that the membrane may be composed of cellulose acetate, polyamide or polyvinylfluoride.

In the Hilton-Davis patent specification or description, no operative hydrostatic pressure other than the range of 200 to 400 p.s.i.g. specified in claim 1 is set forth. As to pH, the specification states (col. 7, lines 55-61) that the reaction mixture fed to the ultrafiltration unit generally has a pH of approximately 9.0, but that it is preferred to adjust the pH to approximately 6.0 to 8.0 before passage through the ultrafiltration membrane.

In the first Office action, the Patent and Trademark Office Examiner rejected all claims of the Hilton-Davis application as unpatentable over Dynapol's Booth et al. U.S. patent no. 4,189,380 which, as you know, discloses a process for purifying polymeric colorants by ultrafiltration through a suitable membrane such as those marketed by Osmonics, Inc. In response to this rejection, Hilton-Davis' attorney amended claim 1 of the application to incorporate the pH range of 6.0 to 9.0 now found in claim 1 of the issued patent and argued that the claimed process patentably distinguished over the Booth et al. patent for the following reasons:

(1) The minimum molecular weight of the polymeric dyes purified by Booth et al. is 610×10^3 whereas the dyes purified by the Hilton-Davis process have molecular weights ranging from 376 to 588.

(2) The Booth et al. process requires the addition of salts to the feed solution whereas the Hilton-Davis process does not require such addition.

(3) Booth et al. teach that it is often advantageous to add and/or maintain additional materials in the ultrafiltration feed to maintain the pH above 9 and preferably from 11 to 13.

(4) Booth et al. carry out ultrafiltration at pressures from 25 to 200 p.s.i.g., preferably at 50-150 p.s.i.g. whereas the Hilton-Davis process is carried out at "much higher" pressures, i.e. 200-400 p.s.i.g., and at much lower pH's, i.e. approximately 9 but preferably 6.0 to 8.0.

After making such amendment to specify the pH range found in claim 1 and presenting such arguments, the Examiner allowed the application and the '746 patent was issued to Hilton-Davis.

Based upon our conference of September 28, 1987, it is our understanding that Warner-Jenkinson plans to commence use of an ultrafiltration or reverse osmosis process using a polysulfone membrane at an operating hydrostatic pressure at least as high as 450 p.s.i.g. and at a pH of 5.5 or below. We further understand that by operating at such higher pressures in excess of 450 p.s.i.g., you obtain the benefit of faster removal of impurities with a resultant higher color concentration (30-35%) in the final product. This in turn renders the spray drying of the color concentrate more energy efficient.

Based upon our above-stated understanding of the operating conditions which Warner-Jenkinson will utilize in its ultrafiltration process, it is our opinion that the practice of such process clearly avoids literal infringement of all claims of the Hilton-Davis '746 patent. Thus, an operating pressure at least as high as 450 p.s.i.g. and a pH of 5.5 or below plainly fall outside the stated ranges set forth in the patent claims.

We have also considered whether or not the practice of the ultrafiltration process under the above-stated operating conditions may subject Warner-Jenkinson to possible liability for infringement under the so-called doctrine of equivalents. Under this doctrine, even though a process may avoid literal infringement of the patent claims, infringement may nevertheless be found if the accused process performs substantially the same function in substantially the same way to achieve the same result. For several reasons, we have concluded that the doctrine of equivalents is inapplicable under the circumstances here obtaining.

First of all, as noted above, Hilton-Davis obtained the allowance of the patent claims only after inserting the recited pH range in the patent claims and arguing that its process distinguished over the Booth et al. patent on the basis of such pH range and the 200-400 p.s.i.g. pressure range. Under these circumstances and taking into account the applicable legal principles, we believe Hilton-Davis is estopped from urging that the stated pH and pressure ranges be "stretched" to encompass the higher pressures and lower pH which Warner-Jenkinson will be employing, i.e. Hilton-Davis will not be heard to say that these parameters (pressure and pH) are material to distinguish over the prior art and immaterial to ensnare an accused infringer. Moreover, the fact that you obtain an added benefit from the use of higher operating pressures also militates against the application of the doctrine of equivalents.

Accordingly, it is our opinion that Warner-Jenkinson by operating an ultrafiltration process under the pressure and pH conditions noted above avoids literal infringement of the '746 patent claims and is also not properly chargeable with infringement under the doctrine of equivalents.

Validity of the '746 Patent

As previously noted, the only prior art reference cited and applied against the Hilton-Davis application which matured into the '746 patent is Booth et al. patent no. 4,189,380. However, in our review of this matter, we have concluded that Osmonics, Inc.'s published March 1978 technical bulletin concerning its SEPA membrane is far more pertinent to the patentability of the '746 subject matter. While this Osmonics bulletin was disclosed to the Patent and Trademark Office by Hilton-Davis' attorney, it was obviously not considered by the Examiner.

The Osmonics bulletin discloses the company's SEPA cellulose acetate polymer membrane and contains a comprehensive teaching of suggested process parameters in carrying out ultrafiltration for different applications (including textile dye removal) and for purifying products of different molecular weights. Thus, for materials with molecular weights in the range 400 to 600 (generally the range noted in Hilton-Davis' arguments to the Patent and Trademark Office), the bulletin recommends membranes with a nominal pore size between 8 and 11 Angstroms and operating pressures between 200 and 400 p.s.i.g. It further mentions the use of pH's between 2 and 8. Interestingly and perhaps not coincidentally, these ranges match or overlap the ranges set forth by Hilton-Davis for membrane pore size, pressure and pH. In fact, the pressure range is precisely that specified in the '746 patent claims.

In our judgment, the disclosure of the Osmonics bulletin is not only clearly more pertinent than that of the Booth et al. patent, but is of such close relevance as to render the validity of the '746 patent claims highly questionable. Applying the statutory standard, it seems clear to us that one skilled in the art having knowledge of the Osmonics bulletin disclosure would regard it as obvious to select a cellulose acetate membrane of a pore size in the range of 5-15 Angstroms and operate at a pressure

of 200 to 400 p.s.i.g. and a pH at least in the range of 2 to 8. The Osmonics bulletin thus contains within its four corners all of the information one skilled in the art would need to develop the patented process.

It is therefore our opinion that there is a strong likelihood that the Hilton-Davis '746 patent is invalid as defining subject matter which was clearly obvious to one of ordinary skill in the art at the time the alleged invention was made in the light of the Osmonics' disclosure.

In summary, it is our considered opinion that Warner-Jenkinson is free to practice its ultrafiltration process at a pressure at least as high as 450 p.s.i.g. and at a pH of 5.5 or below without infringing the Hilton-Davis '746 patent and that the patent is most likely invalid on the basis stated above.

If you or Terry O'Reilly have any questions concerning this matter, kindly let us know. We are retaining copies of the Hilton-Davis file histories, but if you or Terry wish to review them, we will forward copies to you.

Sincerely,

/s/ Don G. Leavitt
DONALD G. LEAVITT

3jmf

cc: Terry O'Reilly

Defendant's Exhibit 632

WARNER-JENKINSON DYE ULTRAFILTRATION

- ▶ Osmonics tested our Red #40 & Yellow #10 dyes at its testing lab in Minneapolis. Aug. 17-19, 1982
- ▶ Analyze Osmonics' preliminary test data. Aug., 1982
- ▶ Received from Osmonics Red #40 & Yellow #10 samples ultrafiltered by Osmonics. Sept., 1982
- ▶ Discussing with Osmonics' technical personnel data performed by Warner-Jenkinson on Red #40 & Yellow #10 process analysis. Oct., 1982
- ▶ Received formal report from Osmonics of August test results on Red #40 & Yellow #10. Nov., 1982
- ▶ Discussions concerning application of ultrafiltration and testing of Yellow #10, Red #40, Blue #1 & Yellow #5 dyes. Nov., 1982 to Feb., 1983
- ▶ Meeting at Warner-Jenkinson with Osmonics' representative and discuss decision to pursue Blue #1 investigation and testing. Feb., 1983
- ▶ Prepare written test objectives for Blue #1 and schedule date for test at Osmonics. Mar., 1983
- ▶ Osmonics performs Blue #1 tests at its testing lab in Minneapolis. May, 1983
- ▶ Analysis of results of tests on Blue #1 and discussions with Osmonics' personnel concerning test results, and problems with chromium molecule, and alter formula for making Blue #1. June-Aug., 1983
- ▶ Schedule second Blue #1 tests at Osmonics. Sept., 1983
- ▶ Formal report by Osmonics of April test results on Red #3. July, 1985

- ▶ Discuss with Osmonics Red #3 test report and analysis and research. Aug.-Sept., 1985
- ▶ Schedule meeting with Osmonics at Warner-Jenkinson to discuss leasing filtration equipment. Oct., 1985
- ▶ Discussion with Osmonics regarding equipment, and investigate alternate supplier of equipment (Pasilac). Nov., 1985
- ▶ Continue discussions of rental of equipment from Osmonics and from Pasilac. Dec., 1985
- ▶ Issue order renting Osmonics equipment and order testing of Red #40 & Yellow #10 on Pasilac equipment. Jan., 1986
- ▶ Osmonics' equipment installed at Warner-Jenkinson, and begin test of Red #40, Yellow #10, Yellow #6, Blue #1, Red #3 and Green #3 with Osmonics' equipment. (Testing continued through July, 1986.) Also, Pasilac performs Red #40 & Yellow #10 testing. Feb., 1986
- ▶ Continue testing. Discuss testing with Osmonics personnel. Mar., 1986
- ▶ Continue with above and second round of tests by Pasilac on Red #40 & Yellow #10. Apr., 1986
- ▶ Successfully manufacture Green #3 without using lead oxide molecule, and discuss Red #40 testing on Carre equipment. May, 1986
- ▶ Successfully manufacture Blue #1 without using chromium molecule, and discuss renting larger filtering unit from Osmonics. June, 1986
- ▶ Osmonics performs second Blue #1 tests at its testing lab in Minneapolis. Oct., 1983
- ▶ Warner-Jenkinson analyzing and discussing with Osmonics the test results. Nov., 1983
- ▶ Received formal Osmonics report on Blue #1 tests. Dec., 1983

- ▶ Analyzing and discussing Blue #1 test results. Jan., 1984
- ▶ Philip Morris to Sell Warner-Jenkinson. Jan., 1984
- ▶ Philip Morris sells Warner-Jenkinson to Universal Foods. Apr., 1984
- ▶ Universal Foods restores research & development funds to Warner-Jenkinson. Oct., 1984
- ▶ Research on manufacturing Blue #1 dye without using chromium molecule. Also begin research on manufacture of Green #3 dye without using lead oxide molecule. Oct., 1984
- ▶ Begin discussions about testing Red #3 at Osmonics. Feb., 1985
- ▶ Continue analysis and order Red #3 testing at Osmonics. Mar., 1985
- ▶ Osmonics performs Red #3 tests at their testing lab in Minneapolis. Apr., 1985
- ▶ Analyze & discussing Red #3 Osmonics test results. May, 1985
- ▶ Continuing extensive research in lab on manufacturing Blue #1 & Green #3 dyes without chromium and lead oxide molecules. June, 1985
- ▶ Order Osmonics' larger filtering rental unit, and visit Carre to view test of Carre equipment on Red #40. July, 1986
- ▶ Second Carre test work on Red #40. Order Pasilac rental equipment. Aug., 1986
- ▶ Discussion with Osmonics regarding start-up of larger Osmonics' rental equipment at Warner-Jenkinson. Aug., 1986
- ▶ Producing full 5,000 lb. batches of Red #40 on Osmonics' larger rental equipment at Warner-Jenkinson. Sept., 1986

- ▶ Selling Red #40 from ultrafiltration to customers of Warner-Jenkinson, and producing Blue #1 & Green #3 on the Osmonics' larger rental equipment, and order Carre rental equipment. Oct., 1986
- ▶ First discover Hilton-Davis' patent. Oct., 1986.

SUPREME COURT OF THE UNITED STATES
OFFICE OF THE CLERK
Washington, D. C. 20543

February 26, 1996

Mr. H. Bartow Farr III
Farr & Taranto
2445 M Street, NW
Washington, DC 20037

Re: Warner-Jenkinson Company, Inc.
v. Hilton Davis Chemical Co.
No. 95-728

Dear Mr. Farr:

The Court today entered the following order in the
above entitled case:

The petition for a writ of certiorari is granted.

Sincerely,

/s/ William K. Suter
WILLIAM K. SUTER
Clerk